Vol. II

TRANSCRIPT OF RECORD

Supreme Court of the United States

OCTOBER TERM, 1941

No. 680

U. S. INDUSTRIAL CHEMICALS, INC., PETITIONER,

vs.

CARBIDE AND CARBON CHEMICALS CORPORATION

ON WRIT OF CERTIORARI TO THE UNITED STATES CIRCUIT COURT OF APPEALS FOR THE FOURTH CIRCUIT

PETITION FOR CERTIORARI FILED OCTOBER 4, 1941.

CERTIORARI GRANTED NOVEMBER 10, 1941.

United States Circuit Court of Appeals

FOR THE FOURTH CIRCUIT

No. 4774

U. S. INDUSTRIAL CHEMICALS, INC.,
Defendant-Appellant

v.

CARBIDE AND CARBON CHEMICALS CORPORATION,
Plaintiff-Appellee

APPEAL FROM THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF MARYLAND

APPENDIX TO PLAINTIFF-APPELLEE'S BRIEF

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United States Circuit Court of Appeals

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No. 4774.

APPENDIX TO PLAINTIFF-APPELLEE'S BRIEF

Testimony of Reston Stevenson

[84]

* * * * *

Thereupon—

RESTON STEVENSON, a witness of lawful age, produced on behalf of the Plaintiff, having been first duly sworn according to law, was examined and testified as follows:

[85]

DIRECT EXAMINATION BY MR. WATSON:

Q. Please state your name, residence and occupation? A. My name, Reston Stevenson. I am professor of chemistry at the college of the City of New York. I reside in New York City, 585 West End Avenue.

Q. What has been your education and experience, Doctor Stevenson? I want your qualifications which enable you to explain the patent in suit.

[86]

A. I was born in Wilmington, North Carolina, in 1882. I graduated at the University of North Carolina, in 1902, with a Master's Degree in Chemistry in 1903; then went to Cornell University for two years as an Assistant Instructor in Chemistry, 1903, -4, and -5, where I also took graduate work in Chemistry. In 1905 to -6 I was Analytical Chemist for the General Chemical Company. I left then to go to the College of the City of New York. And I have been at the College of the City of New York ever since that time. And my position now is that of Professor of Chemistry there.

During the World War, the previous World War, I was commissioned as a captain, and I was detailed to work with the French chemists at the Sorbonne, and elsewhere, upon chemical warfare. A part of that work was upon a catalytic process of making ammonia, by synthesis catalytically from the elements. For this work and for other chemical warfare work that I did with the French I was decorated with the Legion of Honor. I left the Army at the end of the War, but I have recently been appointed a consultant for the Chemical Warfare Service of

[87]

the United States Army.

I have given expert testimony in patent cases; and in three of these patent cases the processes were catalytic.

I am a member of the American Chemical Society, I am a member and Past Sectional Chairman of the Electro-

Chemical Society, and I am a member and Past President of the Chemistry Teachers' Club of New York City.

I have the degree of Ph.D. from Columbia University, which I took in the year 1908 upon Physical Chemistry as a major subject.

Q. Have you read, and do you understand, the patent in suit? A. I have read it, and I do understand it.

Q. Will you please refer to that and discuss the subject matter as disclosed therein, explaining the process, and so much of the general features of chemistry involved as may be necessary to an understanding of it.

The Court: And you may assume, Professor, that you are telling it to a freshman class that knows no chemistry, and therefore knows nothing about this subject.

[88]

A. This Lefort Patent deals with a chemical process in which the gases are caused to react under the influence of a surface catalyst. The gases are molecular oxygen having the formula O_2, signifying two atoms of oxygen drawing together to form a molecule, O_2, such as the oxygen in the atmospheric air; and the other gas that reacts is ethylene.

* * * * *

The ethylene is formula C_2H_4, but the formula given in more detail shows the carbon joined to two hydrogens, the other carbon to two hydrogens, and the double bond between each carbon's atoms, that is ethylene. Ethylene is a gas; and it has a faint odor, and it is inflammable.

The product of the reaction is ethylene oxide, and it will be observed that one oxygen atom is there in addition to the ethylene molecule that was there before. Ethylene oxide is C_2H_4O:

* * * * *

[89]

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Here is a method that is used by chemists to represent this same thing. (Referring to models.) The carbon quite appropriately is black, and the hydrogen, inappropriately, is yellow. But there are others: Two hydrogens, and two more hydrogens. So ethylene is C_2H_4, and there is the double bond between the carbon atoms shown also, whereas in contrast, compare with that ethylene oxide, the red ball meaning the oxygen. And the difference is that one of these bonds has broken out, and has joined on to the oxygen. And the difference in formula is C_2H_4 and C_2H_4O.

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[90]

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The product ethylene oxide is a valuable one, not only in itself, but also as an intermediate for making a number of chemical substances of value. It is the base for making the permanent anti-freeze for automobile radiators. That is to say, the ethylene oxide is the intermediate, because using it as an intermediate it makes glycol; and glycol and water mixed together make the permanent anti-freeze for an automobile radiator, such as the Prestone brand. And this same glycol, with some water, is used as a cooling liquid in airplane engines.

And ethylene oxide is an intermediate for making a number of lacquer solvents. And it is used for the raw materials in diversified industries, such as air-conditioning, safety glass, the finishing of leather and textiles, and also in national defense.

The catalyst involved is a surface catalyst, which is a solid material, of extended surface, which by its presence and its contact with the gases causes and directs the desired chemical combination. The catalyst

[91]

itself takes no part in the chemical combination. That is to say, it takes no part in this sense, that the catalyst remains at the end of the reaction unchanged chemically and unchanged in amount, although it has caused a reaction to take place.

The first two lines of the patent in suit read, "This invention provides a specific and novel process for making ethylene oxide."

In 1931, the chemical compound ethylene oxide was not itself new; but the process is new, and the process is also specific, by which is meant, as will appear, that it makes ethylene oxide directly and without substantial amounts of such other products as would present difficulties of separation.

Lines 2 through 8 refer to the novel process as follows: "It essentially consists in causing ethylene to combine directly with molecular oxygen at temperatures of about 150 degrees to about 400 degrees Centigrade in the presence of a surface catalyst which favors the oxidation of ethylene to ethylene oxide under these conditions."

[92]

I shall refer later to a consideration of these lines.

This oxidation of ethylene to ethylene oxide is a chemical reaction which lines 17 through 20 expresses in symbols as follows:

I shall first read lines 17 through 20. This novel reaction may be expressed as: $2C_2H_4 + O_2 = 2CH_2\text{-}CH_2$ (with O bridging the two CH_2 groups).

Or, as shown here on the chart, this expression, which is called a chemical equation means two molecules of ethylene

plus molecular oxygen form two molecules of ethylene oxide.

The desirability of effecting this reaction was recognized over a century ago by the great French chemist, Berthelot, and he wrote this equation in an article that he published in 1882, and he calculated indirectly, using atomic weights that were prevalent at that time, the heat of reaction if the reaction could be made to occur, but he was careful to say that at his time there was no way of making that reaction occur, but you could start

[93]

with ethylene and arrive at ethylene oxide by a roundabout, indirect process involving several steps.

This inability to effect the direct addition of oxygen to ethylene to make ethylene oxide continued over the years, and in 1926 it was thus expressed in the book of Rideal and Taylor on catalysis at page 213 (reading):

> "The investigations on the direct oxidation by means of oxygen gas of a number of organic substances has indicated that the process can not take place by means of simple addition, for example, 2X plus O_2 gives 2XO, where X is the substance undergoing oxidation to the stable oxide XO."

Expressing this as applied to ethylene, it reads:

> "2 ethylene plus O_2 will not make 2 ethylene oxide."

The patent discloses that this is no longer an impossibility but a fait accompli.

In the equation that we have written here there is no mention of the surface catalyst; because a chemical equation represents chemical changes, for calculating the relative amounts of reactants and products,

[94]

whereas the catalyst does not change in either amount or chemical nature through the reaction; the presence of the catalyst may be indicated by placing the word "catalyst" over the arrow (indicating on chart).

And so we have, then, the two molecules of the ethylene plus the molecule of oxygen, formed in the presence of the catalyst, two molecules of ethylene oxide.

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[100]

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Doctor Stevenson, can you give me an indication of how many compounds of ethylene and oxygen there are?

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[101]

The Witness: If I restrict it for simplicity, starting with one molecule of ethylene reacting with oxygen, there are about seventeen of them that are well known that you could have, could make and they are given on this table here and to make it very brief I will not follow the alphabetical order but will call them off. There is acetylene and oxalic acid and glycolic acid, formaldehyde, glyoxalic acid, alcohol, ethylene oxide, carbon monoxide, carbon dioxide, water, formic acid, acetic acid, dehydroxyethylene, ketene, glyoxal, acetaldehyde, ethylene peroxide, glycolic aldehyde, and those represent what you could get from the oxidation.

By Mr. Watson:

Q. How is ethylene oxide made by this indirect roundabout method you spoke of? A. The indirect method is

shown in this tabulation and is known as the chlorhydrin method in which you have the ethylene react with hypochlorous acid, making ethylene chlorhydrin and afterwards you have the ethylene chlorhydrin reacting with an alkali such as sodium hydroxide and making ethylene oxide. In the equation as written here H_2O has been omitted, it is carried in water.

Q. Is that a catalytic method?

[102]

A. No, that was not a catalytic method, as you can see, because the hypochlorous acid did not remain unchanged chemically and in amount at the end of the reaction. It was changed chemically into the chlorhydrin and afterwards into sodium chloride and as to the amount that was used up and so it was not catalytic and known as purely chemical equation.

Q. Refer to the patent in suit, lines 1 to 8. Is that a catalytic method? A. Lines 1 to 8 I need not read again because I have read them and that states that it is a catalytic method. It mentions the surface catalyst that is there and it also mentions that it is a combination of the surface catalyst and the temperature; the two are mentioned as bringing about the reaction.

* * * * *

[103]

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A. That is a unique reaction in my opinion because so far as I know this was the first time that anyone had taken ethylene and had made that second bond open up and put on one atom of oxygen. In particular, it was the

[104]

first time that that had been done under the influence of a catalyst in particular, but in general I think it is unique also. I think it is somewhat comparable to the adding of hydrogen to ethylene by Sabatier and Senderens, where they put on two hydrogens to make the saturated compound and that is known as the Sabatier Senderens equation as shown in this little tabulation that is published by the Van Nostrand and Company and it has organic type reactions with their originators' names and you find many of the great names of chemistry in this, such as Grignard. You find the Sabatier Senderens reaction there and that is supposed to be sufficiently described there when they say "Passing vapors of substances to be reduced mixed with hydrogen over finely divided nickel or certain other metals (catalyzers) at an optimum temperature.

[105]

A. Ethylene oxide is a liquid but it is a very volatile liquid. It boils at about ten degrees centigrade and this room must be about thirty degrees centigrade, maybe thirty-five, and it is a very volatile, colorless liquid and it has an ethereal odor.

[106]

Its real use is, it is an intermediate, as we say in chemistry, and starting with that you can make many chemicals of value.

[108]

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It makes a number of lacquer solvents. And that is to say, those are called glycol ethers, which means that the ethylene oxide, which is a base, is combined with some organic alcohol. One of those glycol ethers is "Cellosolve."

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[109]

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This patent states that ethylene oxide is formed, and it is a specific reaction. It gives you ethylene oxide; but without the formation of these other substances that would involve difficulty in separation,

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[110]

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Of course, the oxygen of the air is molecular oxygen as it occurs in the air, and the air commonly contains, as well, nitrogen and water vapor, or moisture, and carbon dioxide, and other things.

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[116]

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Ethylene oxide is well known and has been well known for a long time to be perfectly soluble in water, as ammonia gas is perfectly soluble in water. And so it can conveniently be taken out by letting the gases bubble through

water; and that will take out the ethylene oxide, and let the other gases go on.

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[121]

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Q. What does that patent say as to the range of

[122]

temperature to be maintained? A. The patent says that you maintain a temperature in the range of 150 about through 400 degrees Centigrade, about.

Q. And is the reaction exothermic or endothermic? A. Of course, the burning of a hydrocarbon is exothermic, as people know who run automobiles, and that means it gives off heat. And the burning of ethylene, some of which occurs, to some extent, as was shown by this second equation, that is, at line 25, that is exceedingly exothermic, and was known to be so, and the value of it is in the literature before 1931. In fact, as long ago as about 1880, I believe.

And also, the reaction before that, at line 18 of the patent, the equation shown there, that is also exothermic, and that was known to be exothermic and to give off heat, too. And since those are the two reactions that occur, why then, the reaction occurring in the reaction tube is certainly exothermic.

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[123]

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In the first place, water is invariably present, because you have two reactions there, and the side reaction forms

water, and there is water in the reaction tube, where the reaction is going on. So there, at that very spot, there is water as that equation at line 25 shows.

In the second place, water can be present in the air, as in the air today, and moisture in the air; in the third place, water could be actually added, if desired.

[124]

Q. The Court spoke about lines 36, et cetera, about heat control. Were means well known of controlling heat in reactions of this kind, that is to say, how to maintain a temperature within the desired range? A. Yes. Yes, indeed, the catalytic reactions are reactions which are exothermic, give off heat, and there-

[125]

fore when you do them on a large scale you are apt to have so much heat come off that you have to control it, and therefore for the early catalytic reactions prior to 1931, there were a number of expedients known. Of course, the obvious expedient, such as shown by that jacket around the tube, is to have a liquid in there to cool it down sufficiently, not too much, keep it up to the desired temperature but don't let it get too high, and control it, in other words. So, that was known. If you have diluents present, they tend to control the temperature, too.

Now, in regard to the diluents, like nitrogen, oxygen, carbon dioxide, or water or hydrogen, which he speaks of here, which you can add or not, as you prefer, as he states here, why, all of those are diluents, and if you dilute the material it is at once apparent that the rate of formation

of heat is lessened thereby, so that you have less heat to control, so that is one way, and so equalizes the heat there.

However, since diluents are chemicals they can not be indiscriminately added, because some chemicals, of-

[126]

course, will break up a chemical process, and therefore the patent does state certain diluents that can be added.

* * * * *

Q. Now, in line 47 of column 1, it speaks of putting inside the reaction tube a surface catalyst, as one "composed of silver in finely divided form, and which may be disposed within the tube on a supporting material, or in other known ways." What is your understanding of the significance of the words "silver in finely divided form on a supporting material"? A. It mentions silver as a catalyst, and silver as a catalyst is naturally in finely divided form.

As Ellis, in one of his books, states it is so obvious a thing to use a catalyst in a finely divided form that nowadays it seems like an original intuition of the human mind.

At all events, it is customary to have the catalyst finely subdivided, but finely subdivided does not mean that you start with a piece of silver and try to grind it up, which is practically impossible in the

[127]

case of silver, since it is a soft metal, and this has not been ventured by chemists who are skilled in the art. To get finely divided materials they do not grind them up, they would take a compound of the metal, such as a silver nitrate in this case, which would be soluble. Now, to the silver nitrate they add something that would give a pre-

cipitate of silver oxide, the precipitate is already finely subdivided, it is extremely finely subdivided, so that it has been finely subdivided by nature but it still is not silver, so that material would be reduced, the silver oxide would be reduced to silver by means of hydrogen.

The Court: Now, what is the supporting material?

The Witness: The supporting material is a solid which will itself be practically inert and not harmful, and which will hold the finely divided silver on its surface, and the use of the support is to make an economy in the silver, like using silver-plated ware instead of sterling silver, and the foundation can be a piece of brick which has been used successfully, I know, or a piece of porcelain,

[128]

or a piece of Filtros or a piece of Aloxite, and there are a number of materials of that sort that are known, and a number that are in the literature, and in the book of Rideal and Taylor, "Catalysis in Theory and in Practice", published in 1926, on page 123, he mentions the nature of support materials, and he says:

"A variety of substances have been employed as support materials. In addition to those already alluded to, mention may be made of glass, wool, and silica fibres, fibrox, various porous siliceous materials such as fire-clay, lundum, and unglazed porcelain, the various forms of charcoal, anhydrous salts and porous oxides obtained by calcination of suitable materials or by dehydration of gelations, precipitates or gels."

* * * * *

[129]

By Mr. Watson:

Dr. Stevenson, will you refer to claim 8 of the patent? Do you find support for the language used in that claim from the specification?

[130]

A. Claim 8: "The process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportion of one atom of oxygen to one molecule of ethylene"—well, the object of this patent is the production of ethylene oxide, "by the direct chemical combination of oxygen with ethylene", and through my explanation I have emphasized the directness of the reaction, where the oxygen joins directly to the ethylene, and it is shown very thoroughly here in line 18 of the first column, where you see the oxygen joining directly to the ethylene to make ethylene oxide; "in the proportions of one atom of oxygen to one molecule of ethylene."

And looking again at line 18, the equation we see there, that there are two atoms that join two molecules, using those in the proportion of one atom to one molecule, one atom of oxygen for one molecule of ethylene, to make one molecule of ethylene oxide, and the equation is doubled as it is written, each process being a reaction between ethylene and molecular oxygen.

[131]

It is already understood molecular oxygen is O_2, such as in air, and I don't think there is any question whatever

that that is here many times in the patent and we need not take up any time to show it. "Conducting said mixture through the confined reaction zone", that is shown at line 44, the first column, "through a tube, for example, of silica, which is maintained at an elevated temperature". The elevated temperature is shown at 150 to 400 degrees Centigrade; that is on the 5th line of the first column; "Controlling said temperature to maintain said mixture in said zone at a temperature between about 150 degrees and about 400 degrees Centigrade" and then at line 50, it reads: "The temperature of the reaction zone is maintained between about 150 degrees and about 400 degrees Centigrade". For example, at about 200 degrees Centigrade. "Subjecting said mixture in said zone at said elevated controlled temperature—", well, we have gone through that before and there is no use to repeat. "To intimate contact with an active surface catalyst". The active surface catalyst is mentioned at line 4, second column. Then looking at line 47, "Such a surface

[132]

catalyst", line 5, I believe it is in the presence of a surface catalyst, both in the first column, "of a material having the catalytic charactertistics of silver". Now, a material having the catalytic characteristics of silver you will find in the first column, line 48, 47, "Such a surface catalyst is one composed of silver in finely divided form" is one spoken of there. You will find also the statement in the first column, line 5, "In the presence of a surface catalyst which favors the oxidation of ethylene to ethylene oxide under these conditions", and further down at line 15 you find that occurs without the formation of a significant amount of other compound such as aldehydes, so that that shows what is meant by having catalytic characteristics

of silver. Of course, silver itself has this catalytic characteristic if it is silver in finely divided form, a silver catalyst. The next phrase clarifies that statement and reads as I have just said from the specification which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water. I would like to say a word about that since the

[133]

simultaneous side reaction shown at line 25 forms water in the reaction tube, inside the tube, this water is always present when that reaction goes on. That reaction goes on simultaneously with the main reaction shown at line 18 of the first column, so that this is in the presence of water. The amount of water is variable. It can be in air or the air can be dry or the air can be very moist indeed or you can add at will extra water or water vapor or water which becomes water vapor. Now, we have read that phrase: "the presence of water and which does not favor the existence in said product of substantial amounts of aldehydes", and I have already read the part in the patent which related to that when I said it was a specific reaction.

• • • • •

Then the next phrase is "correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form

[134]

substantial amounts of ethylene oxide." Now, that phrase shows that there are three things that go on. One is, that

you pass the mixture through a tube which, of course, you see, and you have in there a catalyst and you have a temperature that is an appropriate one in the given range, that you have. Then, I start with the catalyst. To make it specific, I would prepare a silver catalyst. Now, the patent tells me that I can use it between a range of 150 and 400 degrees and use it with this ethylene and oxygen mixture, preferably ethylene and air mixture, to form ethylene oxide. Therefore, I use that silver catalyst on a support or not in the reaction tube such as you have just seen and I heat it up to 150 degrees and I have the mixture go through at a moderate rate and I note whether I get ethylene oxide substantially or not. It is very simple to note that and then I increase the temperature to say 175 degrees and note again and so by going up at intervals and noting I find that at some place within the prescribed range it will act very well indeed. Then I have correlated the temperature and the catalyst and if I want to I can now change the rate of flow, although that rate of flow is

[135]

not critical, I could use that flow, or many times that amount or half of that amount but there are certain economic considerations which determine what the rate of flow shall be should you get a rate of production where the supply will meet the demand and that would be correlation which is the manner of carrying out the acts or steps as we have already said. It is not a separate step by itself but a manner of carrying out these other steps.

Continuing the last phrase in Claim 8, "And thereafter recovering ethylene oxide-containing product in liquid form." We have lines 17 to 20 of the second column "Ethylene oxide can be recovered by known methods, and it may be further reacted to form ethylene glycol or de-

rivatives of ethylene glycol." Also in the second column, lines 1 to 2, they read "The ethylene oxide can be recovered in any suitable way, preferably in liquid form by condensation or solution." I have now read every phrase in claim 8.

[140]

Q. Dr. Stevenson, we had compared the language of Claim 8 with the specification. And I ask you the general question, whether you thus find the language of Claim 8 of the patent in suit justified by the disclosure of the specification? A. Yes, I find it completely so.

Q. I will ask you the same question as to Claim 9. A. Claim 9 I also find completely disclosed. I need not go through Claim 9 in such detail, because Claim 9 is identical with Claim 8, except at three

[141]

places. And so it seems better that I only mention those three places. One is at line 13. "Heat absorbing diluent." And that is shown on page 1, column 1, line 38, or 37—38. In fact, before that, line 36: "This can be facilitated by suitable dilution of the reaction gases such as that accomplished by the use of air as a source of oxygen and some water or carbon dioxide in addition to that formed can be admitted to the mixture in the reaction zone if desired."

Q. What is the next feature of the claim? A. The next part of the claim which is different from Claim 8 is shown at line 21, which is "Surface catalyst material composed essentially of silver." And this is shown in the patent in suit on page 1, second column, line 4—"The surface cata-

lyst is preferably silver as stated." Also, on that same page and column, line 8 through 11, would seem to be the same thing. "Experiments have also indicated a catalyst may be silver

[142]

activated by the addition of small amounts of gold, or copper or iron," when they are minute amounts, it is still essentially silver.

Then the last part of the claim, the last two lines of the claim read differently from Claim 8, reading, "and thereafter separating ethylene oxide from the products of oxidation."

And referring to the patent, second column, first line, we see that ethylene oxide can be recovered in any suitable way, preferably in liquid form by condensation or solution. And in either case that involves separation from the gases that it is with.

Q. You have spoken of the feature in Claim 9, of the heat-absorbing diluent.

You spoke to me this morning about a word which occurs on page 125 of the transcript, the typewritten transcript. A. Yes, I remember speaking to you and saying that I wanted to make a correction. It is not a cor-

[143]

rection of the stenographer, but is a correction due to my own inadvertence, where I included among the heat-absorbing diluents, and apparently I included hydrogen. I spoke of hydrogen first. And then when I discussed the heat-absorbing diluents I did not specifically exclude the hydrogen. And hydrogen is not a heat-absorbing diluent, since under the conditions of this process it would burn to water and give off heat.

* * * * *

Reston Stevenson

[148]

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Q. What is your opinion as to the relation between the process recited in claims 8 and 9 of the patent in suit and the process of the defendant as recited in the statement just handed you?

• • • • •

A. The defendant's admitted operations as given in this statement conform throughout completely to each and every item in claim 8 and in claim 9. There is a perfect correspondence, and I have, therefore, made this tabulation of claim 8 and claim 9, quoted on one side of the sheet, and on the other side parallel thereto I have quoted extracts from defendant's statement which show the parallelism.

[150]

Q. Doctor Stevenson, would you before concluding give me a summary of what you consider Lefort's contribution? A. In summation I would say that Lefort originated a process in which ethylene oxide is produced directly from molecular ethylene and molecular oxygen by the direct combination of one atom of oxygen with one molecule of ethytene. Lefort discovered that in this production of ethylene oxide he accomplished it by subjecting a mixture containing ethylene and molecular oxygen to his combination of silver surface catalyst and a temperature that is elevated and controlled and is between 150 degrees and 400 degrees Centigrade with water always present in the reaction zone. In Lefort's process the ethylene oxide is produced without substantial formation of aldehydes but with burning some ethylene to water and carbon dioxide.

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[160]

CROSS EXAMINATION BY MR. BARROWS:

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he would use his knowledge as a man who is skilled in catalytic reaction in selecting an appropriate catalytic support or carrier. Naturally, he would require that the catalyst supports do not poison the silver. And naturally he would require that it should not mask the effect of the silver by some catalytic effect of its own. And, of course, he would want the carrier or support to be of a material which would hold the silver, and would hold it spread out. And, apart from that, he has quite a choice of materials that he can take from this page, or elsewhere.

Q. How would he find a support which would be satisfactory? Is there any way except to take it and try it and see?

[161]

A. Yes, for instance, he would see wool fibres. Well, he manifestly would not use wool fibre at a temperature around 300° Centigrade. And you find various other things there. And I know that glass, for instance, is satisfactory. Fireclay is satisfactory. Alundum is satisfactory. Unglazed porcelain. And then you come to charcoal. Charcoal is apt to give a rather dusty product. You would rather not have it. And charcoal, if it is activated carbon particularly, has a catalytic effect of its own. And so you would try naturally something very simple. In fact, the simplest thing that you would try would be silver itself, of course.

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[167]

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The Court: There is no mystery involved in the support used, is there?

The Witness: No, sir, there is not.

The Court: I asked you a question about that, I think yesterday, about the support.

The Witness: Yes. I said that the support is nothing but a solid, mechanical foundation which serves to spread out the catalyst upon it. And that is an economy of the catalyst, especially an economy of a

[168]

silver catalyst. And then I used the analogy of a silver-plated article.

The Court: The reference is on line 48 on the first page, the first column, "and which may be disposed within the tube on a supporting material, or in other known ways."

The Witness: Yes, other known ways, such as pelleting it.

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[177]

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Mr. Barrows: May I ask the direct question of this witness:

Q. Whether he knows any way of designating a silver catalyst other than by the method of preparing it? A. I should say the way it acts, the way it acts, catalytically, and also one may be inferred from the other, but the way

it acts catalytically is certainly a check upon its method of preparation.

Q. And the only way to find that out is to try it? A. Well, I wouldn't say that. If you take the method of preparing silver catalysts by the method of Benton and Elgin which is one that is in the literature before 1931, you will undoubtedly make a silver catalyst by following their method, and it will be active, undoubtedly. I won't guarantee within a degree at what temperature it will be active, no, but it will be active within the range given in the patent.

[178]

Q. Well, do you know any way of telling whether a silver catalyst is effective in carrying out what you consider the process of this Lefort patent except to make it and try it to find out? A. Yes. Yes, you look up the literature on the subject of silver catalysts and you find a number of methods of making them. I have recited one, Benton and Elgin; that will undoubtedly give you a silver catalyst, it will undoubtedly work in this range of 150 to 400 degrees, and it will undoubtedly make ethylene oxide without substantial formation of aldehydes, and you know it in advance.

Q. You say you know that in advance? A. Know that it will?

Mr. Barrows: Yes.

A. Yes, yes.

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[943]

Thereupon—

RESTON STEVENSON, produced on behalf of the Plaintiff in rebuttal, having been first duly sworn according to law, was examined and testified as follows:

DIRECT EXAMINATION BY MR. WATSON:

Q. You have heard the testimony in the case since you were on the stand and gave your testimony in prima facie? A. Yes.

Q. The defendant's witnesses have stated that in their opinion the processes disclosed by the original and Reissue Lefort patents are fundamentally different. Have you read the original patent? A. Yes.

Q. And studied it? A. Yes.

Q. What have you to say in expressing your opinion on the question of fundamental identity or fundamental disparity between the processes disclosed in the original and

[944]

Reissue Lefort patents? A. I found them fundamentally the same.

Q. What are your reasons for that opinion? In answering I may suggest, Doctor Stevenson, that the Court has indicated we want to come down to brass tacks as rapidly as possible, and, therefore, make your answer as comprehensive as you wish but also as terse as possible? A. I think I should first begin in regard to water since his Honor has expressed interest in that particularly, and in the Reissue patent I find that the presence of water in the reaction zone is mandatory and the situation in the reaction zone is a paramount consideration because it is in the re-

action zone that the ethylene oxide is prepared and it is there where any effect of water would be noticed even as any effect of silver in the reaction zone is noticed and besides the two experts of the defendant agreed that consideration of the water should be for the water in the reaction zone.

I find that the Reissue patent makes the presence of water in the reaction zone mandatory in claims 8 and 9. In claim 8 I find at line 34 the phrase "In the presence of water", and that same phrase "in the presence of water" I

[945]

find at line 25 in Claim 9, and obviously "in the presence of water", would mean that water is there and looking at that phrase with more care you see that it is a phrase which defines or describes the active surface catalyst and, that active surface catalyst acts in the presence of water and it is acting while this water is there; also turning to the specification of the Reissue, I find in the first place the equation of complete oxidation at Line 25 and I should like to have the easel brought up with that equation on it, though I shall go on while that is being done.

The equation to complete oxidation shown at line 25 of the Reissue patent shows the formation of carbon dioxide and water and that water that is formed is formed in the reaction zone which is the very place it is needed, if at all, and the equation is the best way to express the fact to a chemist because he understands it very well indeed from an equation and it expresses it quantitatively to him in that equation and in the equation above given at line 18. Water is not a reactant in either equation, although it is a product of the second.

By the Court:

[946]

Q. May I ask you right there, if that is true, what do you understand to be the value of claims 8 and 9? Why do you need those? A. You need claims 8 and 9 because you wanted to have, for instance, a substance essentially silver and not a substance which is either silver or something else or is a metal selected from a group and then you did not want the restriction upon claim 8 and 9 that hydrogen had to be present, which is in some of these claims also.

Q. What did you understand to be Mr. Boynton's stated reason for putting Claims 8 and 9 in? A. Well. I did not listen to him to tell you the truth because I was thinking of what I was going to say. He spoke just then, I really did not listen to him, sorry, but I didn't, but I am willing to give you my reason as I understand it, if you care to hear it.

Q. Very well. A. In Claim 1 we have that the catalyst is silver and must be activated by small quantities of these other materials. That is quite a restriction upon it and you do not want to have that restriction, so that ends that claim.

[947]

Claim 2 is not for silver but is a catalyst which is a mixture, a mixture of silver, gold, copper and iron, so that that is a mixed catalyst and we would rather not have that, at least I do not want to be restricted to that. We do not mind one claim in the patent doing that but we do not want to be restricted.

By Mr. Watson:

Q. Doctor Stevenson, what is the significance of the first equation in the Reissue? A. The first equation of the

Reissue shows that ethylene and oxygen under a catalytic influence will make ethylene oxide and that was certainly the basic pioneer discovery of Lefort and he thought that he was entitled to that, and I presume that Mr. Boynton thought so. I think so anyhow, I will testify, and not to be restricted as some of these claims do.

Q. Referring to that first equation, is that the equation the single equation in the Reissue which shows the production of ethylene oxide? A. Oh, yes, that equation is the equation for the production of ethylene oxide. That is the equation I call

[948]

the Lefort reaction.

Q. What are the reactants as shown by that equation? A. Ethylene and oxygen only and not water and not carbon dioxide.

Q. Turning to the original patent, the original Lefort patent, do you find that same equation in the original? A. Yes, that same equation is at line—well, between lines 10 and 15 on the first page.

Q. Does the equation carry the same significance in the original that it carries in the Reissue? A. It does indeed. It is identically the same equation and means the same thing to the chemist.

Q. What in that case are the reactants yielding ethylene oxide? A. The reactants are ethylene and molecular oxygen.

Q. Refer please to the original patent and tell me whether in your opinion that indicates a permissive use of added amounts of water in the entering gases? A. Why, a chemist would infer from that that water is unnecessary as a reactant for that equation and, in fact,

[949]

if he looked at the other equation that accompanies this as a side reaction, he finds that water is unnecessary as a reactant for that equation also.

Q. Do you find anything else in the original which indicates the permissive nature of the added water in the entering mixture as distinguished from the water in the reaction zone. A. Yes, in a number of places. I find at line 10 on the first page and the second column "Water can be admitted in the reaction vessel", after which I find a comma and therefore I pause and resuming "either in the liquid state, or as steam", and furthermore you will find here—just a moment—"The oxygen can be the oxygen of air." There "can" is used in that sense. I find also in the same sense doubtless "For increasing the yield in ethylene oxide, I am reading from line 19, first page, second column "For increasing the yield in ethylene oxide, the process can also be carried out under a pressure lower than atmospheric pressure", which has no applicability, but I find above that at line 14, "The efficiency of the reaction is increased by diminishing the CO_2 which is formed, by introduction, in this reaction, of a suitable quantity of water." If the efficiency

[950]

is increased, there must be some efficiency before it was increased. Evidently he thought you could at times add water and get beneficial effects.

I find in line 45 of that second column of the first page "If certain quantity of CO_2 is previously introduced", which is permissive or optional, that is for CO_2, in fact, I think no one has questioned that but the same thing is said above with regard to water on line 36, it says "It re-

sults that, if water is introduced in suitable quantity", and so forth. I find on page 2, the second column, lines 1 through 5, "The experiments effected by the applicant have shown that, in presence of the catalyzers indicated, water, in the form of steam or not, considerably promotes the reaction ensuring the production of ethylene oxide". There is no comma after "reaction" and so you read that straight along, "promotes the reaction ensuring the production of ethylene oxide". That is exceedingly clear in the French patent from which it is taken which says: Fr. Pat. 739,562 p. 2, l. 63; "l'eau, sous forme de vapeur ou non, favorise considérable—ment la réaction de production d'oxyde d'éthylène." And so, I admit that I cannot invoke the French because this is not French here, but it took that as a source undoubtedly.

[953]

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In the first place, the Reissue Patent is rewritten, and rewritten in what was considered, and which I consider a better and clearer form. The original patent, however—the Reissue is in the better form, I hope I said—the original patent, however, represented what your Honor called the evolution in the invention that went through the 729952 and the 739562 French patents, with their four additions, and their one addition respect-

[954]

ively, making, therefore, seven of those little French patents. And in those the subject was developed, and he did not develop this second reaction of complete or destructive oxidation until the fourth addition of the 729 Patent, and until the first addition of the 739 Patent. And so I suppose

that in writing them up he wrote them up somewhat in following that order. That is what I think was done. He very clearly says, however, about that, that it is a reaction of oxidation independent from the other reaction; that these two reactions, therefore, are independent of each other; as I call them, side reactions.

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[958]

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The Court: I would like you to give me an answer to that last question. Do you consider that this matter of water is really material, except in connection with the object that the Frenchman speaks about?

The Witness: Well, the Frenchman does not speak about merely omitting the water in the entering gases in that patent. I think you have been misled in that theory, because when it was said that that is the construction that must be put upon that as inventive novelty, the omission of water in the entering gases, why, no reference was made to that very statement, made long before, years before, in resumé 5 of the French patent 729952, and which reads, "By avoiding the addition of water to the reaction one can obtain solely ethylene oxide." So that that could not have been the inventive novelty, because that had already been published, published in France, and years before.

The Court: Well, you just mean he does not know what he is talking about?

[959]

The Witness: No. Well, he may not be right, but he did give a novelty there. He said that he was going

to obtain ethylene oxide without any water present, in the absence of water. In other words, absence of water in the reaction zone, of course, because the British patent that was referred to by Dr. Taylor, who considered those two patents, the French and the British specification together—and they are the same patents, of course—and it reads here—I will quote the very paragraph that he quoted from, page 1, lines 65 to 70, "Thus, according to the present invention, ethylene oxide is produced by the catalytic oxidation of ethylene in the presence of an inert gas and in the absence of water." And I think that is the asserted novelty, because that would have been a novelty at that time. And I have to put upon this patent some construction that would give it life, if true. And that is what I say is the novelty. And, of course, if he did that, that would result in a very great increase in efficiency, none of the ethylene being wasted by burning it up and which would form water, you see.

[960]

Q. (By Mr. Watson) Dr. Stevenson, if any one did operate the process completely in the absence of water in the reaction zone, as indicated in that last British and French patent, do you think that he would be conducting practices covered by Claims 8 and 9 of the reissue patent in suit? A. No, certainly not, certainly not, because those patents, those claims say, "In the presence of water," and this says, as inventive novelty, the very opposite, "in the absence of water," which makes it hard to see how you could invoke a patent requiring the absence of water to put in two claims that read in the presence of water.

The Court: Now, I understand you, but that, I think, puts me squarely up against my first dilemma. You see, the claims require the presence of water.

The Witness: In the reaction zone.

The Court: But the specifications in the reissue patent apparently do not require it in the first reaction.

[962]

The Witness: My explanation of that is that I would not consider it significant because in the specification it is stated that water must be present. In the first place, the fact that it is an accompanying reaction which produces water and goes on at the same time as an original reaction, and I know that it does go on, and I do know that as water is formed more and more ethylene oxide is formed, too; so then, inevitably, water is present, and to the chemist it would mean that, because the equations are to the chemist the most eloquent expression of anything occurring.

I would like to read to you also on the second column of the first page, lines 12 through 15:

> "In any case, the ethylene and oxygen are thus reacted simultaneously at the temperatures set forth in the presence of a surface catalyst and of water."

That certainly reads the presence of water there, so that the specification contains it, and it seems to me to require it at one place rather than another is to criticise it for its literary style but not for the substance of the disclosure.

[965]

By Mr. Watson:

Q. Dr. Stevenson, referring to the original patent and the second equation in column 2, line 40, what produces the Figure 2 H_2O in that equation? In other words, does it come from the reaction of water which has been suggested might be added by him then, or does it come from the ethylene? A. The water in that equation is not any water that was already present but it is water that is formed by a reaction and it comes from the hydrogen atoms of

[966]

ethylene combining with the oxygen atoms of the molecular oxygen.

Q. Well, my point is, considering that second equation, is it true that the preceding mention in the specification of the permissive addition of water to the entering gases refers not to effecting a reaction expressed in the equation, but refers to suppressing the reaction expressed in the equation? A. That is precisely true. At least half of that second column on the first page of the original U. S. is given to the theory that the applicant had in regard to supressing that reaction of destructive oxidation by means of the addition of water, if he cared to do it.

Q. What is your opinion, then, as to whether the side reaction indicated in that second equation in the original, is one which accompanies simultaneously the ethylene oxide formation expressed in that first equation? A. Yes. I know that it does, and I understand that it is so disclosed in this original U. S. patent.

Q. Then what is your opinion on the matter preceding the second equation in the original, viewed from

[967]

the inquiry of whether that matter indicates that the additional water causes this reaction or the contrary? A. No, I know that the addition of water does not cause that reaction. I know that the addition of water is unnecessary to make that reaction shown in the original U. S. at line 41 of the first page, the right hand column, and any addition of water, if made, is made according to the theory of the patentee, to suppress that reaction.

Q. Considering the substance of the disclosure of the original patent, what is your opinion as to its instruction to a chemist that in the production of the ethylene oxide by the equation in lines 10 to 15 of the first column, there is an accompanying side reaction according to the equation expressed in line 41 of the second column? A. The two equations given in the reissue are identical with the two equations given in the original U. S. In both patents they are characterized in such language that the chemist would see that they occur simultaneously and are side reactions. No matter where their place in the patent may be, their place in the

[968]

reaction tube is simultaneous.

The Court: Well, I don't think you need pursue this any further. I understand, in substance, you agree that it should not occur to any one skilled in the art that the addition of water is absolutely required, but that the reaction does take place in the presence of water, whether you put it in there or not?

The Witness: Yes, quite so, water from whatever source, water occurring in the air or deliberately put in or formed in the reaction zone on the job.

By Mr. Watson:

Q. Summarize your opinion, then, as to the correspondence of the original and reissue patents on this question of the presence of water in the reaction zone and the permissive presence of added water in the entering gas mixture. A. I will do that by making a statement first in regard to the reissue, and then making a statement in regard to the original U. S.——.

The Court: Well, you have already done that, haven't you?

[969]

The Witness: Yes, sir, I have, in a sense: I have said for the reissue that the presence of water in the reaction zone is mandatory under claims 8 and 9, and in the specification, and I have read the part of the specification that makes it mandatory. I would like to show that in the reissue patent it is not mandatory for the addition of water to the entering gases, but optional.

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By Mr. Watson:

Q. Have you prepared a tabulation in columns of the disclosure of the original, the reissue, the first French and the second French addition with respect to

[970]

these items of water and things that are in the claim? I just want to have that identified and put it in as an exhibit, because I think it will be helpful. A. Yes, yes, I have prepared this and it refers to the three items given

in paragraphs here, which I gave as the summary of Lefort's contribution; and it has, among other things, water as always present in the reaction zone. That is one of the statements in there.

The Court: Let me have a copy of that.

(Copy of paper referred to handed up to the Court by Mr. Watson.)

Mr. Watson: I think your Honor will find that very helpful. I think I should say, however, that the quotations from the two French patents are given in French, we did not translate them, while there are translations in the case, but they are very simple, indeed, and, I think, it might possibly be more informatory to read them as they are.

[971]

Everything on these sheets are quotations. There is nothing added except the large black print in the lefthand column which is a generalization of the Lefort process. I offer the tabulation compiled by the witness and just referred to by him in evidence as Plaintiff's Exhibit 20.

(Tabulation of Doctor Stevenson marked Plaintiff's Exhibit 20.)

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By Mr. Watson:

Q. Doctor Stevenson, what is your opinion as to the adequacy of disclosure of the original and Reissue Lefort patent with respect to teaching one skilled in the art to produce a silver catalyst effective for carrying on the process?

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[972]

A. * * * I will have to say that I consider the disclosure of both patents is adequate for one skilled in the art, as, in fact, established by the experiments of Doctor Lamb. He got very good efficiency indeed with these experiments. They were efficiencies that correspond and, in fact, were somewhat greater than sometimes is obtained in commercial efficiencies.

[974]

Q. In your opinion, do claims 8 and 9 of the Reissue cover any process not disclosed by the original and indicated by the original to be part of Lefort's asserted invention? A. No.

Q. In your opinion is the recitation in claim 8 and claim 9 respectively of an active surface catalyst of a material having the catalytic characteristics of silver, claim 8, and an active surface catalyst material composed essentially of silver, in your opinion, are either of those definitions of a catalyst functional? A. No.

Mr. Edmonds: It is only part of a definition.

The Court: I did not hear you.

Mr. Edmonds: Counsel asked only about a part of a definition.

The Court: Take it all into account.

The Witness: I will answer it my way and see if it

[975]

answers the question. If I take claim 9 I find that in regard to the catalyst it says "surface catalyst ma-

terial composed essentially of silver." Now, up to that point and not going any further momentarily, that is certainly definite because we know what that means and that is not, I presume, functional at that moment. Now, there is a phrase that follows that, which is a phrase descriptive of the silver catalyst but that phrase that describes the silver catalyst does not in any way make it any broader than it was before and certainly cannot nullify the definiteness that it had before. It has not in any way changed that definiteness, as I understand it.

Now, coming from claim 9 and coming over to claim 8, that same descriptive phrase occurs after the surface catalyst but here the active surface catalyst is of a material which is silver and the materials that are the equivalents for silver in this process as disclosed in the patent from line 4 to line 11 on the first page, second column.

Mr. Watson: You need not quote them.

The Court: I don't think you need to pursue this further because the proof of the pudding is in the eating and

[976]

we have two examples at least that the process is working.

* * * * *

CROSS-EXAMINATION BY MR. BARROWS:

* * * * *

Q. You don't know of any processes where water is added to check the reaction, catalytic reaction? A. Of course, if you added enough water you would check any reaction of oxidation, whether it is liquid water or steam. In fact, you use steam for putting out fires in

[977]

the hold of a vessel but that would be answering it in a general way. I think it should be more specific.

Q. Well, let's be specific. You have mentioned the Benton and Elgin catalyst as a catalyst which you thought might be a proper catalyst to use in the process of this patent. You recall that? A. I mentioned it as a catalyst that I know is good.

[981]

Q. Well, is it your position that this third French patent No. 771650 is one which is supposed to take place in the absence of any added water, and also in the absence of any formed water? A. Yes; yes, indeed. It says the absence of water. I presume it means all water.

[983]

Then, I find that in the third French patent which says very distinctly, and in the British counterpart, that it is to be carried on in the absence of water, and so I take that as what he claims. I do not guarantee that he could accomplish it. But I take it as what he claims. And he also uses the word "suppress" here, suppress or limit it. I suppose that suppression meant complete limitation.

Q. Well, now, Dr. Stevenson, if you assume that the third French patent and the third British patent and the original United States patent are talking about this undesirable

complete reaction to form unwanted products, carbon dioxide and water, and if you assume that that is something to be checked and limited, and only to the extent that you can not prevent obtaining it do you get it,

[984]

and if you think of water which is added afterwards, do you think it is doing violence to English language to construe them as saying that the dry gases he is talking about are the dry gases that go in, and that this unwanted reaction is to be prevented as much as possible— A. Well, I have already explained that he could not possibly have advanced that as his inventive novelty, since it had been published a year or more before. I think it is unnecessary to read that again. I refer to it, though, the resume 5 in the French patent 729952.

Q. But you do agree that the reaction of this third French patent, which results in the formation of carbon dioxide and water is an undesirable reaction, to be checked so far as possible. A. It would be a desirable thing to check; and if you could check it completely, and also have the main reaction going on, of course, it would be a very good invention; I should like to make it myself.

[987]

Q. Well, now, do you find anything in this reissue patent to indicate that this formation of carbon dioxide and water is an undesirable reaction, which is to be checked, and which is to be prevented so far as possible? A. None except the fact that it is obvious, and no man wants to pay

for ethylene and then burn it up to the worthless products we have been talking about.

[988]

Q. Well, is it your position, Dr. Stevenson, that the process of the original Lefort patent, or the process of the reissue patent, is one in which the production of ethylene is by oxidation of ethylene in the presence of a catalyst, with or without inclusion of water in the reaction? A. Oh, no; no, indeed; no, indeed. I made it very clear that the reaction goes on between ethylene and molecular oxygen in the presence of the silver catalyst, and at the temperature range indicated, in the presence

[989]

of water in both patents, in the presence of water in the reaction zone, where the reaction is going on.

Q. Well, then, when Mr. Lefort in his reissue oath referred to something that he wanted to accomplish by his reissue, that is, the production of ethylene oxide by oxidation of ethylene in the presence of a catalyst, with or without the inclusion of water in the reaction, you say that is something that he did not know what he was talking about?

A. I do see something of the sort said by Lefort in the oath, where he wanted—he thought that he had made a disclosure of such pioneer nature that it ought to include the reaction between the ethylene and molecular oxygen, even if some one later arrived at suppressing that reaction of complete oxidation which results in the formation

of water. He had not arrived at suppressing that himself. But if

[990]

any one in the future did, he made that statement there, because of the pioneer nature of his invention. And I understand a good deal of latitude is given to pioneer inventions. He did say, however, later—he made that later in the subsequent oath—he did not claim so wide a scope. He saw that his file history had been such that he could not claim any such thing; and he also realized, perhaps, that the fact that he had not yet really achieved it would not permit him to reach out that far, because later he did not, he did not in the later patent state that.

[992]

Q. With a large amount of steam present blanketing the oxygen molecules or atoms on the surface of the silver catalyst, you would restrain the complete oxidation of ethylene to carbon dioxide and water? A. Well, I don't know what you mean by a large amount. If you would put a very large amount of steam in, neither one of these reactions would occur. Of course, I have said you put out fires in the holds of ships by putting in steam under pressure, so it would certainly stop this.

[993]

Q. Now, you said in your direct examination, comparing the original and the reissue patents, that the equation,

that is, ethylene plus oxygen to give ethylene oxide is the eloquent thing to a chemist. Now, in catalytic chemistry where you have a surface with ethylene and oxygen and steam molecules on the surface would you say that it is also of significance if you have a large enough number of steam molecules on the surface to retard or restrain complete oxidation of ethylene to the undesirable or unwanted reaction product? A. No, I think not, in a practical way, for this reason: I know you can remove nearly all the water so that you have almost nearly, say, two one-thousandths of a per cent of water, in the entering gases, and in that case you will get a production of ethylene oxide from the catalytic reaction of ethylene and oxygen over silver in the range specified here and get good efficiency. I know that if you increase that by adding a little steam, which is the popular term for water vapor, water vapor

[994]

being the generic term, if you add a little water vapor then you will still get the production, you can still add the water vapor. I have had experiments performed up to 3½ per cent water vapor, and in that range I got about the same efficiency. There was practically very little difference among them. Why should there be? It is the difference between a moist—not between a dry reaction and a wet reaction—it is the difference between an inevitably moist reaction and one which is a little moister. It is true that I did not keep the temperature constant, no I raised the temperature until in each case I was getting the best results, because, naturally, I compared each one acting well with the other.

[1001]

The Court: Dr. Stevenson, I have one question: As far as the French patent, that is, the late patent, which it has been suggested is the reason for the introduction of the reissue claim——

A. Yes, sir.

[1002]

Q. Am I correct in this understanding, that what you claim for the French patent is that it is an attempt to make ethylene oxide completely without water? A. Indeed, that is what I think is the asserted novelty.

Q. And therefore, if I understand you correctly, you say that whether we assumed that it was considered and known by those who made the Reissue application, that as you interpret the Reissue claims, they are entirely distinct from the French patent in that you find that water is still mandatory in the Reissue claims? A. Yes, that is true, sir.

Q. In the new claims of the Reissue patent? A. Yes.

Q. And that regardless of whether the French inventor has a successful operating process, regardless of whether his patent is valid or invalid, that you contend that the two things in claim under that third French patent and the claims under the new claims of the Reissue patent are entirely distinct? A. That is exactly my attitude, exactly.

[1006]

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Mr. Watson: The Plaintiff offers in evidence a copy of the Taylor patent heretofore marked for identification. * * * Exhibit 10.

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[1007]

(Exhibit previously marked Plaintiff's Exhibit 10 for Identification now marked Plaintiff's Exhibit 10.)

Mr. Watson: I offer Plaintiff's Exhibit 21, a folder of correspondence and cables which has been supplied us by defendant's counsel and therefore the authenticity of which I presume will be accepted.

[741]

Testimony of Emory P. Boynton.

Thereupon——

EMORY P. BOYNTON, a witness of lawful age, produced on behalf of the Defendant, having been first duly sworn according to law, was examined and testified as follows:

DIRECT EXAMINATION BY MR. EDMONDS:

[743]

Q. Now, you are familiar with this cablegram, which was marked as Exhibit No. 5, aren't you (handing paper to witness)? A. Yes, sir.

Q. Was anything received from Mr. Chalas or his associates, sent by him or them, between March 6, 1936,

[744]

and the date of this cablegram, March 12th? A. No. As I recall the facts, and I think I know them, the cablegram

which we received on March 12th was the first communication which we received from Mr. Chalas.

Q. And then this Exhibit No. 3 came in next? A. Yes.

Q. And what was the next thing, this Exhibit of March 28th, this cable of March 28th? A. No, there were some other exchanges of cables between our company and Mr. Chalas, dealing with the matter of how soon he could get a definite order, arrange with the French company for——

* * * * *

Q. The actual consummation of the purchase of these patents was a little later, wasn't it? Or, rather, this

[745]

one patent we are talking about, and possibly something else. That came a little later, didn't it, in May or June? A. No, you have a cablegram there saying that the deal has been closed, or words to that effect.

Q. Yes. A. Dated March 28th.

Q. I mean the actual signing up? A. Yes. And we requested a lawyer from Norway to come to Paris, and he did, and drew up an agreement, which was then merely a protocol agreement.

Q. Do you remember the date when the deal was consummated? A. About the middle of April, 1936.

Q. And was there more correspondence prior to that date, or cables, than what you have already referred to? A. Between which dates?

Q. Up to that date of closing, April 16th, or whatever it was? A. Yes, but they were mostly with this lawyer in Norway, about how soon he could get down to Paris to draw

[746]

the agreement.

* * * * *

CROSS-EXAMINATION BY MR. WATSON:

Q. Did any of these other cables or correspondence that Mr. Edmonds referred to deal with the chemical subject matter, or did they deal with financial terms? A. Only with terms.

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[747]

* * * * *

Mr. Watson: There is one question I want to ask. The Court will recall that the occasion for the witness was to allow Mr. Edmonds to find out whether there was a document in existence. Now, that question Mr. Edmonds did not ask the witness at all.

Q. (By Mr. Watson) Did you make a search for such a document? A. I presume that you refer now to the hypothetical

[748]

or possible United States application corresponding to British specification 431966 and the French patent 771-something?

Q. Yes, sir. A. Yes, I searched the assignment records in the United States Patent Office, under the French company, Francon and Lefort. I also asked the French company if they had any more applications than the one application which they assigned to us with the original Lefort patent. I also asked the United States attorneys for the French company whether there was another application than that one. I did everything that I knew how to do to find its existence. And I found none.

* * * * *

[796]

Testimony of George H. Law.

Thereupon——

GEORGE H. LAW, a witness of lawful age, produced on behalf of the Plaintiff in rebuttal, having been first duly sworn, was examined and testified as follows:

DIRECT EXAMINATION BY MR. JOHNSON:

* * * * *

[812]

* * * * *

Q. Did you recently conduct some informatory tests at the South Charleston Laboratory in the presence of Doctor Frazer and the representatives of the defendant? A. Yes, we did.

Q. Have you the notebook for that? Are these the ones? A. Yes.

[813]

Mr. Johnson: We offer the notebooks as physical exhibits.

(Second and third notebooks of Doctor Law marked Plaintiff's Exhibits 15-A and 15-B.)

Q. Can you give us a blueprint of the apparatus which was used in that experiment? A. Yes, I can.

Q. Is that it? A. That is it.

Mr. Johnson: We offer the blueprint identified by the witness as Plaintiff's Exhibit 16.)

(Blueprint of apparatus identified by witness Law marked Plaintiff's Exhibit 16.)

* * * * *

[840]

Thereupon—

GEORGE H. LAW, recalled by the Plaintiff, testified further as follows:

REDIRECT EXAMINATION BY MR. JOHNSON:

Q. Doctor Law, will you explain somewhat more in detail what you did in South Charleston in the presence of Doctor Frazer and the representatives of the defendant? A. We prepared a silver catalyst by precipitating silver oxide from a solution of silver nitrate. We washed that carefully to remove any impurities, made a slurry of it in water and evaporated it to dryness in the presence of a Filtros carrier or Filtros support, which is an artificial form of silica. That was placed in a glass tube practically identical to that.

Mr. Johnson: Indicating Exhibit 3.

The Witness: And so reduced with hydrogen for four hours over a temperature range of 100 to 250 degrees. We then started passing the mixture of air and ethylene over the catalyst. We found originally when we started with about a five per cent. ethylene and 95% air at a feed rate of about

[841]

50 litres per hour, our first attempt at operating this, we had difficulty in controlling the temperature due to the rather crude apparatus we had. We wanted a glass apparatus in which everything would be perfectly obvious and clear. This was not considered as a test. It was more

of a demonstration. We allowed the temperature to drop somewhat and then started feeding up more ethylene to the rate of about two or three per cent. and got a positive test for ethylene oxide and a negative one on the inlet gas and I am not able to recall exactly the efficiency on this.

[842]

As I recall, we got about a 10 per cent yield, and something around a 30 per cent efficiency. We did not attempt to operate that for any extensive period of time. It was, as I say, merely a demonstration of how we would prepare a catalyst. At the time Dr. Frazer arrived we had an oil-jacketed tube in operation, which contained a catalyst which had been made in exactly the same way, the same support. That had been operating, as I recall, for about 48 hours when he arrived. We attempted to isolate some ethylene oxide from effluent gas that that gave. But the apparatus we used was admittedly not enough to get a quantitative recovery of ethylene oxide. However, we did recover some, and demonstrated that it was present. I think that—yes, in the entering gas we had a water concentration of 1½ per cent. The feed rate varied somewhat from 50 litres an hour, as I recall, to somewhat lower. The ethylene concentrations also varied. I believe we varied them from about 5 per cent to 15 or 20 per cent. We continued operating that tube for an additional 48 hours, and raised our yields to about 32 per cent, and efficiencies of the order of 60 per cent.

[843]

Q. Can you tell me whether or not it has been your experience with catalysts, properly prepared, they nevertheless increase in yield and efficiency of ethylene oxide production as they are run? A. Yes, that is usually the case.

Q. In the reaction zone in those experiments that you conducted, take, for example, the one in the glass tube, where no additional water was added to the air-ethylene mixture being fed in, can you tell us what the condition was as to the water in the reaction zone?

* * * * *

A. As soon as the reacting gases hit the catalyst, ethylene oxide, and water, and carbon dioxide would be produced simultaneously, as nearly as we can determine, and the production of both ethylene oxide, and water, and carbon dioxide would continue throughout the length of the catalyst bed.

[844]

By Mr. Johnson:

Q. And so far as the chemistry of the process is concerned, what is the effect of changing the inlet water content, for example, from two-thousandths of a per cent to two thousand times that amount? A. There would be no difference in the chemistry of the process.

* * * * *

Cross-examination by Mr. Barrows:

* * * * *

[846]

* * * * *

The Court: As I understand it, Mr. Law, you are in substance saying that you agree with Professor Lamb, that you do not think the use of water makes any difference one way or the other.

The Witness: Chemically, no, it is the same reaction.

The Court: Do your experiments, culminating in this particular experiment, lead you to the same conclusion that I understand Professor Lamb testified to, that the use of water is not a material variation in the process?

The Witness: That is correct.

The Court: That you can or can not use it at will?

The Witness: That is right.

The Court: And that you will get the same, substantially the same result?

The Witness: That is correct.

* * * * *

[851]

* * * * *

Q. Well, you do know that this catalyst without added water to the catalyst will not operate at that temperature, don't you? A. No. I have operated catalysts of this identical composition on another support, over the entire range from 150 to 400 degrees, in an oil jacketed tube.

[861]

By Mr. Barrows:

Q. One other question, Doctor Law. Before you undertook this work early in 1936, you had had a good deal of experience on catalytic oxidation reactions, I take it? A. Yes.

Q. Including the catalytic oxidation of ethylene? A. I don't know whether I had done any work on that or not.

* * * * *

REDIRECT EXAMINATION BY MR. JOHNSON:

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[862]

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Q. Your attention was called to U. S. Patent 2142948. May I ask whether, if you know, your company is using that process of activating silver surface catalysts commercially? A. No, they are not.

Q. Were there other well known ways of activating silver surface catalysts than that? A. Yes, there were.

Q. Prior to 1931?

[863]

A. Yes.

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[890]

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Testimony of Granville A. Perkins.

Thereupon——

GRANVILLE A. PERKINS, a witness of lawful age, produced on behalf of the Plaintiff in rebuttal, having been first duly sworn, was examined and testified as follows:

DIRECT EXAMINATION BY MR. WATSON:

Q. What is your occupation, Dr. Perkins? A. I am employed in the research and development department of the Carbide and Carbon Chemicals Corporation, as a chemist.

Q. How long have you occupied that position?

[891]

A. I have occupied that position since 1929; eleven years.

• • • • •

Q. Can you give a brief statement of your qualifications as a chemist? A. I graduated from Cornell in 1913 with the degree of A. B., specializing in chemistry, and have the degree of Ph. D. from the University of Pittsburgh, in 1917.

I have been in various positions since then in chemistry, and spent eight years in the Philippine Islands, working for the Philippine Government in the research and manufacture of drugs for the treatment of leprosy. And then in 1929 I was employed by the research and development department of the Carbide and Carbon Chemicals Corporation, and since then have been engaged in chemical work.

Q. You were present during the demonstrations made in South Charleston before Dr. Frazer? A. Yes, sir.

Q. You have been present during the trial?

[892]

A. Yes, sir.

Q. It has been suggested by a witness for the defendant, who was also present at that demonstration, that there might have been a feature that was unusual or abnormal in the selection of Filtros as the carrier on which to deposit the silver catalyst. What have you to say on that? A. Well, I see nothing unusual in Filtros as a carrier. I have known it used commercially as a carrier since 1929 for catalytic operations commercially. It is spoken of in a patent which issued in 1914, as a carrier; and, furthermore, it is silica in an inert form. Now, we have heard considerable about silica gel here as being an inert carrier. The fact is that silica gel is the most active form of silica that I know of; and it has been used as a catalyst itself in a great many operations. And it is also well known that any substance when finely divided, or in a form having a greatly extended surface, such as silica gel, is very active catalytically as compared with the fused substances. Now,

Filtros is silica gel in a fused form, or, at least a form which has been heated

[893]

quite highly, and is one of the most inactive forms of silica that I know of.

* * * * * *

Q. What have you to say on the possible criticism of the demonstration, that you were unwarranted, or that it was something unusual to use as much as 30 per cent of deposited silver on the support? A. Well, commercially catalysts are used on carriers sometimes in small amounts, sometimes in medium amounts, and very often the catalyst is put in the reaction zone without any supports. That is, 100 per cent catalyst, comprising the contact mass, as it is called. And we have

[894]

tried silver in percentages of anywhere from around 2 to 100 per cent of the catalyst mass; and it works in all of those percentages. I believe that Mr. Carter's records show the same, as nearly as I can recall; that is, that he tried several percentages, starting in with about 2, 50 and 100 per cent, and that usually he got ethylene oxide with those percentages, provided he did not poison the silver with something that might be there. So I think there is nothing unusual commercially or otherwise in the choice of thirty some per cent of silver. We had to take some per cent.

Q. You heard the testimony of the defendant's witness, Mr. De Jong? A. Yes.

Q. Who recited what he did in Emoryville, in California? A. Yes.

Q. What have you to say to his ability and the propriety of his laboratory technique or procedure on the question of its constituting a proper inquiry into the operativeness of the Lefort disclosure?

[895]

A. Well, Mr. De Jong at the time of the experiments certainly was an inexperienced laboratory assistant. He had only made a few tests, I believe about two hours on each test; and I would say that these series of tests were in the hands of an unskilled man, one entirely inadequate to test any catalytic process. All catalytic processes, or, at least, most of them, are rather delicate as far as manipulation is concerned, and one must be careful not to allow traces of poisons to influence the catalyst.

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[896]

Q. Had you finished your answer? A. Well, I think we can say also that from the instructions that Dr. Groll gave Mr. De Jong, that the proposition had not been very well considered by Dr. Groll, assuming that he was a skilled man, because no really good silver surface finely divided catalyst was tried in that whole series of experiments. Silver turnings are all right for high temperature operations, particularly if they are worked long enough so that they become activated on the surface, if they acquire the characteristics which I think Mr. Carter described of his rivets. But to take fresh silver turnings, as Mr. De Jong did under Dr. Groll's instructions, and to try them for two hours, and then put a little mercury on, and also work a short time to try to get the mercury off, mercury being a powerful catalyst poison, I do not think it showed very good technique.

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[897]

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CROSS-EXAMINATION BY MR. BARROWS:

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Q. Do you think the treatment of silver with mercury and distilling off the mercury is not a proper method of making a silver surface catalyst? A. I think it is a proper method if it is worked long enough so that you can be sure that all the mercury is off the

[898]

silver, but if the mercury is not all off the silver, it would tend to poison the surface.

Q. Are you saying that in the test that was carried out the mercury was not all off the surface or don't you know? A. The data we have, I don't think shows convincingly that it was or was not, but I am inclined to think that it was not since it did not give the desired results.

* * * * *

[922]

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Mr. Watson: Before we adjourn, I have asked Mr. Edmonds to consent to or admit that that photo copy of the form of assignment used by the defendant is the usual standard form of such assignment.

Mr. Edmonds: That is the common form used by the Air Reduction Company, Incorporated;

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[924]

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"AND I HEREBY further covenant and agree that I will communicate to the said Company, its successors, legal representatives and assigns, any facts known to me respecting said improvements, and testify in any legal proceeding, sign all lawful papers, execute all divisional,

[925]

continuing and reissue applications, make all rightful oaths, and generally do everything possible to aid the said Company, its successors, legal representatives and assigns, to obtain and enforce proper patent protection for said improvements in all countries."

• • • • •

Testimony of George O. Curme, Jr.

Thereupon—

GEORGE O. CURME, JR., a witness of lawful age, produced on behalf of the Plaintiff in rebuttal, having been first duly sworn, was examined and testified as follows:

DIRECT EXAMINATION BY MR. WATSON:

Q. What is your position and occupation, Doctor Curme? A. Vice President and Director of Research for the Carbide and Carbon Chemicals Corporation.

Q. Are you a chemist? A. I am.

Q. Please indicate briefly some of the honors which you have received in recognition of your ability as a

[864]

chemist? A. I received the degree of Bachelor of Science at Northwestern University in 1909. I received the degree of Doctor of Philosophy in Chemistry from the University of Chicago in 1913. I was a Fellow at the Mellon Institute for the period from 1914 to 1920. I have been employed by the Carbide and Carbon Chemicals Corporation since its formation in 1920 to the present time.

Q. I think we can sum it up, I am sure the defendant will not object on the ground of a leading question because it will perhaps be less embarrassing to you. Your accomplishments as a chemist have been recognized, have they not, among other ways in the tribute of American Industry given this past winter, giving medals to the outstanding American Pioneers in Chemistry? A. That is correct.

Q. And you were included as one of the nineteen recipients for such an honor? A. That is correct.

Q. Doctor Curme, were you advised of Doctor Laws' February 1936 tests in passing ethylene and air over a silver catalyst to produce ethylene oxide, were you advised of those at the

[865]

time? A. I was.

Q. What conclusion did you draw from them with respect to the process under inquiry?

* * * * *

The Witness: I drew the conclusion that it was a

[866]

distinctly operable process and one of great commercial value.

By Mr. Watson:

Q. What action did you take at that time? A. I proceeded immediately to get in touch with the president of our company and my associates in New York and we at once began negotiations for the purchase of the American rights to this patent.

Q. Those negotiations were consummated in an understanding whereby you obtained them in March, 1936? A. That is correct.

Q. What did your company do with respect to the process from that time until you built your plant? A. We began immediately a program of development work to adapt this process to an engineering embodiment which would operate satisfactorily under the conditions existing in our West Virginia plant.

Q. What are these photographs which I hand you? A. These are photographs of our commercial units taken shortly after its completion, which completion date was in November, 1937. I notice these photographs are dated December 21, 1937.

[867]

Q. Has the plant been in operation since then? A. The plant has been in uninterrupted operation since then. We have doubled the size of its operation by engineering changes at one time which required a short interruption of operation.

Q. Can you tell us generally speaking whether the erection and design of such a plant involves a considerable amount of engineering? A. It does indeed. One of the important parts of my responsibility is the operation of the development department of the Carbide and Carbon Chemicals Corporation, the purpose of which is to adapt processes which we have invented in our research depart-

ment or which have been acquired from outside inventors to an engineering embodiment that can operate successfully on a large scale basis.

Mr. Watson: We offer these two photographs in evidence as Plaintiff's Exhibits 17 and 18.

(Two photographs of West Virginia plant marked Plaintiff's Exhibits 17 and 18.)

Q. You have spoken of your duties in the development

[868]

and translation of processes from the laboratory to large scale industrial production. Can you tell me in the light of that experience how the interval between March, 1936 and the commencement of the operation of the plant in November, 1937 compares with other instances of your development and engineering experience to put processes into large scale operation?

• • • • • •

A. This is the most rapid development and adaptation to commercial practice which our company has ever experienced.

• • • • • •

Q. You have been in the Court room during the trial? A. I have.

[869]

Q. I understood Doctor Reyerson to express the opinion that a process including the passage of ethylene and air, the air having added to it, water, irrespective of its amount, and passing that mixture over a catalyst of silver to produce ethylene oxide was chemically speaking entirely different from a process of passing ethylene and air over the

same catalyst to produce the same product. Do you agree or disagree with that opinion? A. I disagree with that opinion. To me, the two are identical.

• • • • •

Q. From a chemical viewpoint? A. That is right.

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CROSS-EXAMINATION BY MR. BARROWS:

Q. On this last question, Doctor Curme, do you know what takes place at the surface of the catalyst when you

[870]

have present ethylene, oxygen and a large proportion of steam? A. From the standpoint of actual knowledge, no one knows. There are many engaging theories on catalysts and a great deal of valuable work on that subject is being done but if you are asking from the standpoint of absolute knowledge, I am sure that there is no absolute knowledge on this subject.

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[1250]

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Thereupon—

GEORGE O. CURME, JR. resumed the stand on behalf of the Defendant, and testified further as follows:

DIRECT EXAMINATION BY MR. BARROWS:

Q. Doctor, do you recall that in this affidavit which you filed, or which was filed in the Patent Office in connection

with Lefort Reissue Application, you referred to the Willstatter and Bommer article, and the work which was done under your supervision shortly after that article appeared. A. I did.

Q. Now, I read from your testimony in the pre-trial deposition, and ask you if that is your testimony now. Page 42.

"Q. Do you recall what temperatures were used in those experiments carried out under your direction

[1251]

after the publication of the Willstatter and Bommer article? A. No, I do not recall except I feel very sure we were employing substantially the same temperatures as were recorded in the Willstatter and Bommer article.

"Q. In your affidavit you said that 'Willstatter and Bommer reported that oxidation catalysts in general lower the temperature at which effective oxidation of ethylene with molecular oxygen takes place.' Is that something which was confirmed by the experiments carried out under your direction? A. Yes.

"Q. Then you say in your affidavit: 'The lower reaction temperature would be expected to be favorable especially because some of the oxidation catalysts were known to be effective in producing formaldehyde by the oxidation of methanol.' What were the oxidation catalysts that you were referring to as catalysts known to be effective in producing formaldehyde by the oxidation of methanol? A. Copper and silver have been used rather

[1252]

extensively in the production of formaldehyde from methanol.

"Q. Copper and silver were well known oxidation catalysts at that time? A. Yes.

"Q. And I believe you said that when you used silver and copper in those experiments carried out under your direction, after the Willstatter and Bommer publication, that you failed to get formaldehyde, or did you state what your results were? Did you obtain formaldehyde when you used copper and silver following the Willstatter publication? A. The only statement I recall is that we obtained substantially the same results as Willstatter, and Willstatter did obtain formaldehyde from ethylene with certain of his catalysts, and we did also."

Mr. Watson: There is a correction in the transcript there, Mr. Barrows. Put in an "if" before "Willstatter".

Q. (By Mr. Barrows) "And if Willstatter did obtain formaldehyde from ethylene with certain of his

[1253]

catalysts, we did also.

"Q. You say you did also? A. Yes.

"Q. And do you recall that you obtained it with silver? A. I would say at this time I can not recall which experiments were conducted at that time. I can only say that we did not discover ethylene oxide in our reaction products or any of the experiments which we conducted."

[1254]

"Q. Did you look for ethylene oxide in the products of that process? A. Not specifically, because we had no reason to anticipate that it would be there. However, we

had, for the time, good research facilities. My brother was a well-qualified organic chemist, and I feel sure that he would have recognized it had it been there in substantial quantities."

Is that still your testimony, Dr. Curme? A. Yes, as mentioned previously, when considered in the light of the various definitions and considerations that had been established earlier in the deposition.

* * * * *

[1257]

Q. Do you recall any work that had been done under your direction in the laboratory of the plaintiff company in the use of different kinds of silver as an oxidation catalyst prior to this work? A. I previously referred to work done in the early 20's by my brother, H. R. Curme, in which he reported some work of Willstatter's in which silver was attempted as a catalyst. Silver had been abundantly mentioned in the chemical literature as a catalyst, and I feel sure that in our investigations on catalytic problems, silver must have been included among the materials examined.

"Q. Do you recall what kind of a silver catalyst your members used in the experiments on the Willstatter process? A. No, I do not recall the details of these experiments.

"Q. Do you recall any experiments in the use of silver gauze as a catalyst prior to these experiments that were discussed this forenoon being the first five experiments in Notebook, Defendant's Exhibit 8 for Identification?"

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[1258]

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Q. That is still your testimony is it, Dr. Curme?

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A. Why, yes, that is my testimony, which is to be taken in consideration with the conclusions which were reached that Willstatter taught us nothing regarding oxidation under the conditions described by Lefort.

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[1274]

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Thereupon——

GEORGE O. CURME, recalled by the Plaintiff, testified further as follows:

[1275]

DIRECT EXAMINATION BY MR. WATSON:

Q. You have heard the testimony, Doctor Curme about the McElroy patents? A. I have.

Q. Does your company own those patents? A. They do or they did up to the date of their expiration.

Q. When did you buy them? A. 1922.

Q. How many did you buy at the time? A. They were purchased with a group of some twenty odd patents acquired from the Commercial Research Corporation.

Q. Did you pay any substantial amount for them? A. The whole group of patents were acquired at a very low figure.

Q. Did they ever teach you how to produce ethylene oxide from that day forward? A. They did not. Technically, they were among our assets. We studied them to find out what, if any, their

[1276]

objects were and we considered them as completely valueless as regards teaching us anything about the manufacture of ethylene oxide without the use of chlorine.

Mr. Watson: That is all.

Mr. Edmonds: May I ask you to wait just a minute.

CROSS-EXAMINATION BY MR. EDMONDS:

Q. That is what you thought about the Lefort patents when you saw them, that they were inoperable, isn't it? A. Until we started to experiment.

The Court: That is all.

(Testimony of the witness concluded.)

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[834]

Testimony of J. C. W. Frazer.

Thereupon—

J. C. W. FRAZER, a witness of lawful age, called by the Court, having been first duly sworn, was examined and testified as follows:

DIRECT EXAMINATION BY THE COURT:

Q. Dr. Frazer, did you witness any tests by either one or both of the parties since the Order of the Court? A. I witnessed——

Q. I do not mean for you to give me or tell me what they were, or the results, but just answer the question, did you witness any? A. I did, I witnessed one series of tests.
Q. Run where? A. In Charleston, West Virginia.

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[835]

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Q. (By the Court) Were representatives of both companies present? A. Yes, sir.
Q. And then, did you consider that was sufficient for your purposes in order to testify here? A. I considered it representative, yes,

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Q. (By the Court) They were run by whom, in your presence? A. They were run by the plaintiff.

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[1036]

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DR. J. C. W. FRAZER (recalled), a witness heretofore sworn, was recalled by the Plaintiff, examined and testified as follows:

Mr. Watson: Preliminarily, may I make two comments? I thought I indicated last night I would be glad to call Dr. Frazer. The second observation is that I really did not understand your Honor's order as you now indicate it with respect to our examination of Dr. Frazer, because that order, I thought, recited that the parties should put in their evidence, then Dr.

Frazer would be called by the Court and cross-examined by counsel for both sides. But, faint heart never won fair lady.

DIRECT EXAMINATION BY MR. WATSON:

[1037]

Q. Dr. Frazer, you have attended the trial? A. I have.

Q. You have heard the testimony? A. I have.

Q. In the course of the evidence a difference of opinion has arisen between the witnesses on the two sides with respect, generally, to two topics; the first one, as I understand it, is as to the adequacy of disclosure of the original and reissue Lefort U. S. patents for the purpose of teaching one skilled in the art how to make a silver catalyst which could be placed in a confined reaction zone and over which could be passed a mixture of ethylene and air to produce ethylene oxide, without the formation of substantial amounts of aldehydes. What is your opinion on that subject?

First, may I qualify you a little more on it by saying that you also attended a demonstration in South Charleston in plaintiff's plant? A. I did, as I said yesterday afternoon.

Q. (Mr. Watson) Oh, yes. And I should have included in my prior question that this process was to be

[1038]

conducted at a temperature within 150 to 400 degrees in the reaction zone. That was a further qualification. Now, what are your views on that subject? A. Well, the question, as you state it briefly, is what would I do to prepare a silver catalyst that would work according to the directions as I would find them in the patents?

Mr. Watson: That is a paraphrase of it.

A. Well, I would proceed exactly, I think, as Professor Lamb did, or, at least, my mental operation would be that.

I would assume that an active silver catalyst was needed, a highly active silver catalyst. Now, I know that there are cases where a catalyst may be too active; one of those is in the catalytic oxidation of ammonia, but I certainly would not consider that here in the first instance.

I would think that I needed a very active catalyst, and I know from all of the work that I have done on catalysts that that material would have to be finely divided in order to have this catalytic activity I spoke of.

[1039]

Now, I know that the best way to get silver in a finely divided condition would be to precipitate some insoluble salt of silver from which catalytic silver could be readily obtained after it was dried or cut into proper shape.

Now, two of those compounds have been mentioned here, two of the three that I would think of at once: silver oxide, which was the starting point in the preparation of the catalyst at the demonstration at South Charleston; and silver oxalate, which has been mentioned in connection with another patent spoken of in this case; a third compound which I would think of, would have been the carbonate.

Now, any of those three compounds will enable you to prepare silver in a very finely divided form.

[1040]

In the case of the oxalate and the case of the oxide, as Professor Lamb carried it out, you can go directly by heat to the metallic silver but at Charleston this was done by reduction in hydrogen. Now, just stepping aside for a minute, that method of using the oxalate has been pat-

ented, I think, if I am correct, and that is a very common thing. One can precipitate as oxalate any metal whose oxide is readily reduced and by careful heat go from the oxalate to the finely divided metal. That is done by most students in the laboratory. They prepare various oxalates, dry them, heat them; they go to a finely divided iron which takes fire when exposed to the air and is called pyrophoric iron. Nickel is the same way and it has been used as a means of preparing nickel catalysts in finely divided condition and the same is true of the silver which is the most easily reduced oxide of any of these metals, so that to come back to where I started, this would be one of those and I would not be able to make any choice in the beginning. I think any of them would work, would be a starting point for the preparation of finely divided metallic silver which I would know and would be perfectly sure would be required. Does that answer

[1041]

the question?

Mr. Watson: Yes, I think that answers the question.

By Mr. Watson:

Q. You speak of the use of the silver compound oxalate and said as I understand you, that its reduction to silver was familiar routine to students? A. Yes, you find it in the text books.

Q. How long has that been rudimentary common knowledge? A. That is an old observation. I don't know how long it goes back.

Q. Does it go back ten years? A. Longer than that, I think it would go back——

Mr. Edmonds: It is enough if you say more than twenty years.

The Witness: Yes.

Mr. Edmonds: You need not strain yourself.

The Court: Wouldn't we get along a lot faster and better if Doctor Frazer told us what he witnessed and then whether he thinks what was done follows the teachings of the

[1042]

patent, and whether the patent was sufficiently descriptive to enable those that perform the experiment to get good results? That is what we want?

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By Mr. Watson:

Q. You heard the testimony of Doctor Law describing the demonstration in South Charleston? A. I did.

Q. Just for brevity of record, it is not necessary to repeat too much of that unless you wish to express some further details about it, but I would be interested in yielding you the floor entirely, Doctor Frazer, and let us hear the opinions which you hold with respect to that operation and with respect to the adequacy of disclosure of the

[1043]

patent and with respect to this question of variance between the original and Reissue? A. I think, your Honor, that you really do want to know about these tests at Charleston, so I will go into those.

Q. Yes, if you please. Will it help you if we bring up the model? A. I think it will save time probably. We

arrived in Charleston, I think, about eleven o'clock or a little before eleven at least.

The Court: Would you talk a little louder.

The Witness: When we arrived in Charleston, we went directly to the laboratory on the morning of the 29th of May. When we arrived we were told that an experiment was already in operation and it had been in operation a matter of a few days, four days, as I remember the number. That experiment, the apparatus used, we were informed, was essentially in its details a duplicate of this apparatus here with the exception of two points; one of those was that the tube on this first apparatus which was in operation, the tube which contained the catalyst itself was metal and not

[1044]

glass, and this metal tube was immersed directly in a heating fluid for better temperature control.

I jotted down some figures that were given on the analyses there during the course of this experiment and possibly could be identified in the sheets that have been submitted of these tests that were made there.

In this first apparatus, the first analysis, I have here was on a sample of gas which contained 19.9% of ethylene and so on. The catalyst bed was at a temperature of 226°. The exit gases contained 7.6% of CO_2 and 15.4% of ethylene and the calculated efficiency was given to be 34%. The second run some time later was made on a sample of gas containing less ethylene. In this gas the incoming gas contained 10.1% of ethylene and the exit gases contained 5.3% of CO_2, 5.8% of ethylene and the temperature was 235 and the efficiency was about 37%. Now, that apparatus was then connected up so that the ethylene which was being generated could be frozen or collected by refrigeration.

Mr. Watson: You mean the ethylene oxide?

The Witness: The ethylene oxide, I beg your pardon, so that the ethylene oxide which was being made in the course of this experiment could be liquified and examined at a later

[1045]

time of the day. That was done for that purpose. Then, this last apparatus was put into use and we were shown exactly how this catalyst was prepared. In fact, the catalyst was prepared there before us. A certain amount of this material Filtros was weighed out, a certain amount of silver nitrate was weighed out and dissolved. The silver was precipitated with a slight excess of caustic soda and washed twice, I think, with water. After the silver oxide had settled from the solution, the solution was poured off, decanted. That was done, I have the number of times, but it must have been ten times, it was continued until the wash water which was being poured off was no longer alkaline and showed the absence of the small excess of caustic soda which was the test for complete washing of the precipitate. That was a preparation, we might say, a suspension of the silver oxide. That was then poured into the weighed sample of Filtros in a porcelain dish, placed on a heater, slowly evaporated and stirred in the meanwhile so as to be sure that this silver oxide when it was dried would be on the surface of the Filtros.

The figures I have as given here before for that

[1046]

particular catalyst would have contained 32 % of silver. That catalyst prepared in that way, after drying, was put in a tube, either this tube or one just like it by placing some

of the untreated Filtros on the bottom so as to have all of the parts that contain the catalysts at a reaction temperature and then the catalyst was poured on top and the tube filled up with a blanket of Filtros just as it is there and then instead of decomposing the oxide by heat alone, as Professor Lamb did yesterday, hydrogen was passed through that tube. It took about three and a half hours as I remember, to reduce completely that oxide. The temperature was 200, or a little above at the end of which time the hot zone which marks the place where hydrogen is reduced to oxide could be followed gradually down through the catalyst bed by moving the thermo-couple junction, and when the heat zone passed out the bottom the catalyst was completely reduced. Two runs were made on that catalyst. The first at 185 degrees, when the inlet gas contained 2.1% ethylene, the exit gases contained 1% CO_2 and 1.3% ethylene, and I was given the efficiency as calculated to be 31.5%.

The second run on that same catalyst was made at a temperature of 179 degrees and on which the inlet gases had

[1047]

4.9% ethylene, the exit gases 4% ethylene and 1.2% CO_2. I was given a calculated efficiency for that experiment of 38% and at this time, that was pretty late in the day, this first experiment which had been allowed to run and accumulate the ethylene oxide was opened and while there wasn't a large sample there, it was ample to demonstrate that ethylene oxide had been prepared. We all examined it, two cubic centimeters.

By the Court:

Q. What was the conversion rate of that, do you know? A. It was part of the test that had been going

on, as I gave the first two analyses that were taken from that run as it was going at the time when we arrived.

Q. I mean, would you say it was commercially satisfactory or not, applying the same— A. As far as I know the technical requirements, I should say yes.

Q. Applying the same percentage test to Professor Lamb's experiment and the others, what was the conversion? A. The efficiency was 34% and 37% on these two experiments.

[1048]

Q. Was the result as efficient in the experiment under the large operation as it was under this specially prepared experiment? A. These two experiments were practically the same scale.

Q. They were? A. Yes, that is practically, I was told, we did not see it because it was in operation in an iron container and I was told this was a duplicate in essential details of that apparatus. The only difference was that the first one was made of iron and this is of glass so you could see it.

By Mr. Watson:

Q. And you could see that the height was about the same? A. I might say one other difference. In this glass apparatus the catalyst tube is not immersed directly in the heating liquid but there was a space of, an annular space of a few millimeters of air in between, which prevented satisfactory temperature control.

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[1052]

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Q. Before the recess we were speaking of some figures. During the recess did you get those data? A. I have these data as given in Professor Lamb's report, and as given in the sheets that are in evidence here on the two experiments on the Charleston run, the day we were there at the laboratory.

Q. Yes, sir. The Court inquired as to what those data were. A. Well, these data, your Honor, represent the percentage of ethylene which is converted to ethylene oxide in the passage through the catalyst tube. They are the percentages of ethylene converted into ethylene oxide during the passage of the gases through the catalyst

[1053]

tube. And I find that those figures for the experiment which Professor Lamb reported are on the average quite a little above the corresponding figures in the Charleston test, both of them.

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[1064]

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By the Court:

Q. Then, wouldn't it have been possible within a day or two after my order was signed for each side to have had a model and to have had Doctor Frazer come to wherever the model was and run the gases through it and show him how it worked?

Mr. Fairbank: We did so within nine days after your order was signed.

The Witness: This order was on the 21st, I received it on the 21st.

The Court: You did substantially that for the plaintiff. The plaintiff did substantially that, but the defendant did not. Is that what I am given to understand?

The Witness: That is correct.

. . . . .

[1073]

. . . . . .

By the Court:

Q. You have studied the patents, haven't you, doctor?

[1074]

A. Yes, I have.

Q. I would like you to give us your opinion as to the use of water, whether it is mandatory or discretionary in the Reissue patent?

[1075]

And in any event, whether you find that there is any material departure in respect to water or anything else in the reissue patent, from the original patent.

The Witness: Well, your Honor, as far as I could make out in regard to this question of water, it is a very much over-worked topic.

The Court: It is what?

The Witness: It is a very much over-worked topic. I can go through the whole thing, all the patents, and indicate what they have meant to me, if that is any help.

Q. (By Mr. Watson) What do you mean by it being an over-worked topic, Dr. Fraser? A. Well, it seems to me that the original French patents and their additions disclose sufficient information in regard to the effect of water, to show that the operation could be performed either with or without the deliberate addition of water to the entering gases.

In claim 5—not claim, but in Item 5 of the original of the French patent—and, remember, that that patent

[1076]

was taken out—

Mr. Fairbank: —729, your Honor, is the first one.

The Witness: That patent was taken out for the production of ethylene glycol. And in that operation you have the two steps performed in one; that is, the oxidation of ethylene and the combination with water.

Now, in order to carry out those two steps in one you must have water. But he says in Item 5 of that that if you omit water you can produce ethylene oxide alone. So that it seems to me that it goes right back to the original patent, that the use of water for the production of ethylene oxide would be considered optional.

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[1082]

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The Court: Dr. Frazer, do you want to say anything else about this water content?

The Witness: Well, aside from the two main reacting gases, which are ethylene and oxygen, there are three other gases, you might say, that have been dis-

[1083]

cussed. One of them is nitrogen.

Now, so far as this operation goes, nitrogen remains inert. It takes no part chemically in the reaction at all. All it is, it is a heat absorber, and tends to prevent explosion.

Now, when you come to carbon dioxide, that also is a diluent, and absorbs heat and prevents explosion; but it is one of the products of the undesirable reaction. And it has, in effect, a third effect: It tends to supress this undesirable reaction. That is one of the reasons why it is advantageous, I should say, to use CO_2 as the diluent rather than nitrogen, because it cuts down the amount of ethylene that is wasted by complete oxidation.

[1084]

When you come to water, as I see it, water does all that carbon dioxide does, but there must be a little water in the reactants to start that reaction, because if the statement in the patent is true—I have never tried it—if the gases are entirely dry, you get no reaction; so that I say that the evidence is that there must be a little water, and that amount is somewhere

between zero and twenty or twenty-five millimeters vapor pressure that you have in ordinary air.

[1102]

By Mr. Edmonds:

Q. In that third French patent he practically told the world that what he had been saying up to that point was necessary, is no longer necessary because of another in-

[1103]

vention he has made, that is true, isn't it? A. Which he had already made in 1931.

The Court: Gentlemen, this doesn't seem to me to be getting anywhere.

The Witness: I think it is haggling over words.

[1108]

Mr. Watson: Dr. Frazer, in the test demonstrated to you in South Charleston, it is my understanding that in the steel tube the entering gas mixture had a water vapor content of about 1½ per cent, and in the demonstration with the pyrex glass equipment the water vapor was about .4 per cent, about 4/10 of one per cent. Does that assist you in expressing an opinion on the effect of differences in the water vapor content?

The Witness: Well, Mr. Watson, my opinion is that the amount of water beyond a certain small critical amount would not make very much difference.

Mr. Watson: And you base that opinion upon what you saw in South Charleston, the consideration you have given to the patents, and your general experience?

The Witness: It is more on what I know about chemistry.

[1109]

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The Court: You mean whether you get it through the air or put it in.

The Witness: It does not make any difference where you get it. And beyond that amount I do not think it would have made a great deal of difference what you do, because once you get it started you get water formed by the second reaction, which adds on to whatever you put in.

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[1111]

By Mr. Edmonds:

Q. Let me ask you this: Have you had time here since last Monday, when the papers were produced, to find from the papers brought in here by the Dutch Shell people who practiced this process in association with the French people, what amounts of water they put in? A. No, I do not recall that. It is unfortunate that I did not get much of Mr. Pyzel's testimony. There was much I could not hear.

Q. Well, it is largely in the documents that were filed here. A. I told you that this morning.

Q. So you have had no opportunity to see that these documents show that the people who practiced that inven-

tion, under the instruction of the French people, put in water vapor up to more than sixty per cent of the total volume.

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Well, all I can say is I don't see why they did it.

[1117]

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We are dealing here with ethylene oxide which, to the chemist, is a somewhat peculiar molecule, because it is what we say is a strained molecule, to form this three-numbered ring, and for that reason it is unstable to the extent that it is strained in its formation and it may easily go to aldehyde or something else.

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[1120]

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The Court: I might as well state now, in order that you may know the way my mind is running, it seems to me that unless the defendant's experts can bring out, can give me a clearer illustration than they have done, or can convince me more than they have done, it seems to me that the weight of the testimony indicates what Dr. Frazer and Professor Lamb and Professor Stevenson have said, namely: that the question of water is a secondary one, and that to use it or not to use it is not a material departure; that in order to be successful in the experiment you probably have always got some water, and the amount of how much is a matter of pure experimentation. That's the way my mind is running now. If that is true, I should have

to hold that the reissue is a perfectly valid patent and is a clarification of the original patent.

The one other question being the question of the catalyst, I think it self-evident from the testimony here that there is a sufficient description.

. . . . . .

[1124]

By Mr. Edmonds:

Q. Doctor, I believe there is some further statement on this subject that you feel that you can make that will be helpful to the Court. Will you proceed with it? A. The testimony that I gave yesterday on this matter of validity was practically confined entirely to the question of water, and I either said or implied that in my judgment the amount of water or the manner in which it was added was not the important thing at all, speaking entirely as a chemist.

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[1133]

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By the Court:

Q. From the chemist's point of view, Doctor Frazer, what is the real difference between giving the wide

[1134]

range of catalysts which apparently are so well known to the art, what is the difference between doing that and providing an optional method of applying heat to a given process? A. Well, catalysts in many cases are very specific and this happens to be one of those cases where the catalyst chooses a reaction going in one direction.

Q. That is true, but it tells you what kind of catalyst in broad terms, it must have the catalytic characteristics of silver. Suppose that was said to be built in a certain way in order to get this desired heat applied in a certain way and then it gives you, however, certain alternate ways of building it providing you produce a certain temperature at a certain time or something like that, as a practical matter, what is the difference as long as silver catalysts are so well known? A. Well, in the future it may not be so. It may be something else that is discovered which will do this thing that is not silver.

Q. And yet has the characteristics of silver? A. Of this particular reaction, the characteristics.

Q. In other words, your real objection is to the

[1135]

phrase "having the catalytic characteristics of silver." You think someone may come along and find something that has no relation to silver but will have the characteristics of silver to produce this effect. A. In this reaction.

Q. You don't think that that argument is true of the language employed in claim 9 because there it has a composition of silver? A. There it is definitely stated to be silver.

* * * * *

By Mr. Watson:

Q. What you said, Doctor Frazer, is a bridge to be

[1136]

crossed when we get to it. Is that your point, some time in the future, the question may arise? A. It means building a new bridge.

Q. You mean maybe building a new bridge in the future? A. Yes.

Q. It is not a bridge that is already built? A. No, it is another bridge.

Q. So that inasmuch as the bridge will be built in the future, we will cross it, if at all, in the future? A. That is correct.

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[1137]

Q. When we want to judge the matter of equivalence between silver and something that somebody may or may not find in the future, we will want at that time to have the evidence on the relationships of the two in all its chemical and practical phases, wouldn't we? A. We would have to.

Q. And likewise, the possible objection you have raised as to that language in claim 8 would not apply if that language is to be interpreted by the Court as meaning materials referred to in the specification of the patent itself, isn't that true? A. That is a right long question.

Q. Yes, I know it is. My point is if as a matter of law, the language in that claim is to indicate a surface catalyst of a material having the catalytic characteristics of silver, if as a matter of law that covers only the materials in the disclosure of the specification, then your objection would not apply, isn't that true? For example, osmium and thalium are not mentioned in the specification of the patent? A. Yes.

[1139]

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Q. Considering the specification of the reissue, you agree, do you not, that it has been narrowed with respect to the number of catalysts recited? A. That is the fact, yes.

Q. Well, now, if, as a matter of law, that language in Claim 8 as to material having the catalytic characteristics of silver was held to mean the materials listed, as the Court pointed out in lines 47 and 48 of column 1, and lines 1 to 11 of column 2, then this

[1140]

possible objection which you have raised would not be applicable; isn't that true? A. If that is the interpretation, yes, because that is the way it appears in claim 9, it is silver.

Q. It seems so to me also. You were asked as to how you would have proceeded in making a catalyst, and you said you would have proceeded about as Dr. Lamb did. A. I think I mentioned three silver salts, that I think either one would work.

Q. And I think you mentioned that you would have transformed those salts into silver, either decomposing them by heat, or reducing them by hydrogen. A. I would have reduced by hydrogen.

Q. But both of those ways were old, common and well known, were they not? A. That is right.

Q. For more than ten years? A. That is right.

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[1143]

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Q. And you agree, do you not, that in the original United States patent it is disclosed to have water present in the reaction zone? A. Yes.

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[1149]

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By Mr. Barrows:

Q. Have you any explanation of why it is that that large result was obtained at Charleston and the small result was obtained by Dr. Lamb, using silver on the different carriers? A. Mr. Barrows, I think I said yesterday that the

[1150]

nature of the ethylene oxide would make it necessary for any one to look more carefully into the nature of the support than would ordinarily be the case, because, as I said, ethylene oxide is, to the chemist, or should be, a somewhat unstable molecule and it will not stand contact with any kind of a surface under the conditions in this catalytic chamber, so I think a neutral carrier would have to include its neutrality toward ethylene oxide or it would not survive in the experiment.

[1266]

The Court: Well, I would like to ask Dr. Frazer a question or two about that.

Thereupon—

J. C. W. Frazer (recalled), a witness heretofore sworn, was recalled by the Court and

[1267]

questioned as follows:

By the Court:

Q You have heard the testimony of Professor Taylor; do you agree or disagree with him with respect to the anticipatory character of the McElroy patents with relation to the patent in suit?

[1268]

A. I think I disagree with him.

Q. For what reason? A. Well, for one reason, these patents were taken out in 1918 and as far as I know there is no evidence that they have ever been used.

Q. Do you consider they are an insufficient disclosure to do what the patent in suit calls for? A. I don't think they are commercial operations.

Q. Do you mean to say that if you took the McElroy patent, either or both of them, that you could not work out a successful process for making ethylene oxide with a silver catalyst? A. That would be my answer.

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By Mr. Edmonds:

[1272]

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Q. Now, if you were practicing this McElroy process or attempting to do it, what is it that you would be lacking in your effort to practice it in the way of directions from McElroy which would embarrass you in your attempts to practice the process which you are told in the Lefort specification? A. Well, I think the two things that would rather stump me, to begin with, is the fact that he doesn't tell which of this class of labile oxides may be useful. In the second place he doesn't tell anything about the temperature range within which such a catalyst would be useful and it would probably be true that the temperature range would be different according to the particular one of these labile oxides you might select, so it would be a complicated thing to work out a combination that would give——

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[253]

Testimony of Lloyd H. Reyerson.

Thereupon—

LLOYD H. REYERSON, a witness of lawful age, produced on behalf of the Defendant, having been first duly sworn according to law, was examined and testified as follows:

[254]

DIRECT EXAMINATION BY MR. EDMONDS:

Q. Will you state your name and place of residence and occupation?

[255]

A. Lloyd H. Reyerson, my residence is Saint Paul, Minnesota. I am professor of Chemistry at the University of Minnesota.

Q. How long have you served in that capacity? A. Twenty-one years.

Q. Have you had occasion to give much thought and study to catalysts? A. I have spent a great deal of time studying catalysts. As a matter of fact, practically all of my research activity since I received my doctorate at Johns Hopkins University in 1920 has been devoted to the study of the effect of surfaces upon gases of various kinds.

[257]

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Q. Now, the statement also is that the process is one of subjecting ethylene to the action, among other things, of oxygen of air. What can you say about that with respect to the amount of moisture content in it? A. Well, air is composed of a mixture of oxygen, nitrogen, inert gases, some carbon dioxide, and a variable quantity of water vapor.

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[260]

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The Court: The application of water is a pretty common thing, isn't it, in a lot of processes. Is it necessary to a trained chemist, to tell how much water he should add to the process?

The Witness: No, I do not think it is essential at all, [to tell] how much water.

The Court: Then, would you say that the fact that the amount of water is not specified would make any difference?

The Witness: It is not a material difference.

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[265]

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By Mr. Edmonds:

Q. What is the purpose of the process described in this patent? Is it the purpose of producing ethylene oxide? A. Yes.

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[278]

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Q. Will you state what is said there about the process to which this Reissue patent was directed? A. The second sentence of the first paragraph states, "It essentially consists in causing ethylene to combine directly with molecular oxygen at temperatures of about 150 degrees to about 400 degrees Centigrade in the presence of a surface catalyst which favors the oxidation of ethylene to ethylene oxide under these conditions." There is no mention of water in that particular paragraph and it mentions molecular oxygen, which in my opinion, means you could use dry oxygen. It does not have to come from the air. It could be the separate oxygen in which case the water content is but negligible; in other words, the statement does not require the presence of water.

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[286]

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Mr. Edmonds: And that the list of catalysts has been cut down from some astronomical figure to something very modest.

The Court: Now, what do you think is the effect of that? Suppose he had an option of catalysts, does that make much difference?

The Witness: Oh, it makes a tremendous difference.

[287]

The Court: Well, you have an option in both.

The Witness: Yes.

Lloyd H. Reyerson

The Court: But the option is simply very much smaller in the second one.

The Witness: That is correct.

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The Court: Well, now, that is evidently what has been attempted in boiling the number down in the claims.

The Witness: That is true.

The Court: Or, as a practical matter, that is a help rather than a hindrance, isn't it?

The Witness: Yes.

[300]

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The Court: But assuming that somewhere in that list you get your catalyst, as I understand it you think with that qualification the first patent does give you a workable specification?

The Witness: Yes.

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[690]

CROSS-EXAMINATION BY MR. WATSON:

[694]

Q. Since you studied either the original or the reissue patent, have you tried to make a silver catalyst for conducting the process? A. No.

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[697]

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Q. • • • you testify that the amount of water as far as the Lefort patent is concerned is not significant nor material. Now, do you mean that in your experimental work in 1926, however, the amount of water as distinguished from the presence of water would have been a very important and

[698]

essential and material factor? A. I think it would with our catalyst, yes.

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Q. • • • will you agree with me, as Doctor Taylor agreed that for all purposes of this question of water, we may confine ourselves to the condition in the reaction zone? A. Yes, if all of these conditions are considered in the reaction zone.

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[701]

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Q. Do you know whether there was published at the date of the issuance of the original Lefort patent that it was published knowledge that silica gel would transform ethylene oxide to acetaldehyde at such temperatures? A. I don't recall any such statement of the literature that silica gel itself would do that.

Q. If that should prove to be the case, it would indicate that silica gel was not a desirable inert support to be used in this process, isn't that true? A. Yes, but until it is proven that, I would not accept the statement that silica gel was not a good carrier.

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Q. It was not silver merely, your invention concerned platinum and in general, its subject matter was

[702]

metalized silica gel, isn't that true? A. That is correct.

Q. And you solicited a patent on that particular substance? A. Correct, that is correct.

Q. Do you know of any commercial catalytic process of oxidizing hydrocarbons where that catalyst of yours has ever been industrially used? A. No.

Q. Will you agree with me and with the prior witness who stated that in 1935, say, when the original Lefort patent issued, the literature indicated a variety of suitable inert supports that might be employed in such a process? A. Yes.

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[703]

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Q. Will you agree with the prior witness that it would have been common sense to select a support which had the necessary qualities of mechanical strength and surface to which the silver could adhere? A. Yes, I would.

Q. Now, I come to the question of your testimony on variance between the original and reissue patent and first, you went over the disclosure of the original patent as to water. I am not going to take you over in detail on that but I want to ask you whether for all the purposes of the cross-examination and for all the purposes of the

[704]

issues of this case, we may not, both with respect to the original patent and the reissue, we may not confine our

consideration on the subject of water to the ambit of the reaction zone? A. That is what you must confine it to.

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[705]

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Q. • • • I will ask you the question whether, in your opinion, the process can be conducted by the addition of water to the entering gas

[706]

mixture so that in the reaction zone all the ethylene oxidized is converted exclusively to ethylene oxide. A. I do not believe so.

Q. Have you the copy of the original patent in front of you? A. Yes.

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Q. Well, then, let us continue down on the same page to line 48, where he says, "the reaction of oxidation of ethylene into carbon dioxide will be completely checked." Those are the same words which you have just used. Do you think he was mistaken in that?

[707]

A. He was mistaken, but that actually applies also to the addition of carbon dioxide.

Q. And that language is omitted from the reissue, isn't it? A. As far as I know.

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[709]

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Q. Then, I go back to my question, and ask whether this omission in the reissue is not, in your opinion, as a practical thing, an improvement, beneficial? A. Well, I might admit that the elimination from the reissue of the addition of carbon dioxide to completely check the reaction might be considered as some addition, some simplification.

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[717]

Q. Dr. Reyerson, have you seen the operations as conducted on this process in the defendant's plant? A. Yes.

Q. How much ethylene oxide is there in the exit gas from the reaction zone, in per cent?

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[718]

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A. Well, I visited the plant in December of 1939; and I am frank to confess I don't remember the percentages now that were in the exit gas.

Q. The percentage of ethylene oxide in the exit gas relative to the percentage of water vapor in the exit gas did not impress itself on your mind as a matter of importance? A. Not at that time.

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[719]

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Q. Well, don't you think the words "considerably promotes" connotes that the reaction occurs even without that

water added? A. No, I would not necessarily feel that way.

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[720]

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Q. Take the next sentence, beginning at line 12, there it says "Oxygen can be oxygen of the air." Doesn't that indicate that the oxygen can be pure oxygen? A. Yes, it just tells you that you may add oxygen of the air. You must add oxygen.

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[723]

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Q. By the way, why did you omit reference in your direct examination to the 5th resume of the very first filed French convention patent 729952? I think your Honor might wish to look at that 5th resume? A. Would you state the question again?

Q. My question is why you omitted reference to that fifth resume in all of your direct examination? A. Because when you take the second addition of this same patent, it states that tests made by the applicant show that the catalyst indicated did not enable one to obtain ethylene oxide by the oxidation of ethylene if the gases are dry. In the first place, I was not asked about this statement. In the second place, I would consider in the light of this additional material it was pure speculation.

Q. Read into the record that fifth resume, please? A. "By avoiding the addition of water in the reaction one can obtain ethylene oxide".

Q. The subject of that first French patent was the production of ethylene glycol, wasn't it? A. That is correct.

[724]

Q. And that involved a combination between water and ethylene oxide? A. Yes.

Q. So that this fifth resume indicates that by avoiding the addition of water the glycol formation might not occur but that the ethylene oxide formation would occur, is that true? A. That is the speculation of it.

Q. That is the import of the statement, isn't it? A. Yes,

* * * * *

Q. Now, the second addition to which you refer is

[725]

epitomized, isn't it, in the statement on page two of the translation "The presence of water is necessary for the production of ethylene oxide"? A. Well, that is certainly part of it.

Q. And we have already agreed that that means that the presence of water is necessary in the reaction zone? A. Is that right? The presence of water is necessary in the reacting gases.

Q. And the gases react in the reaction zone? A. They react on the catalyst.

Q. And the catalyst is in the reaction zone? A. Yes.

Q. Turning again to the original United States Patent, in the first column an equation is given, is that right? A. That is right.

Q. It appears from that, doesn't it, that in the formation of ethylene oxide, carbon dioxide is not a reactant with ethylene to yield ethylene oxide? A. Carbon dioxide?

Q. Yes. A. I don't see anything concerning carbon dioxide

[726]

reacting.

Q. And is it not true that from this equation it indicates that carbon dioxide is not one of the reactants? A. Carbon dioxide doesn't, according to this equation react into ethylene.

Q. And according to that equation, water does not react with the ethylene to give ethylene oxide, is that right? A. That is correct,

[727]

Q. In your direct examination I understood you to state that Claims 8 and 9 of the Reissue begin that the catalyst must be such as will not work in the presence of water. That is not just what you meant?. A. I am sure I never said that. At least, I would like to see that.

Q. Certainly. At least, that is not your opinion? I quote that answer from page 302 of the record. "In other words, the two, that is the original and Reissue, are different because the first requires water and the second does not, but you admit that the second may be a workable plan". That was the Court's question and your answer was "Yes, the first one requires catalysts which will operate in the presence of water and the second one requires catalysts that won't operate in the presence of water." A. Yes, because water is permissive in the second one and if it is to be operated without water, you must have a catalyst that will operate without water.

Q. But inasmuch as you say water is permissive, then in a case where that permission is availed of, it is not true that the claims require a catalyst that will not oper-

[728]

ate in the presence of water, isn't that right? A. It requires a catalyst that will not operate in the presence of water but may be operated in the presence of water.

* * * * *

Q. Turning again to the original United States patent on page 1, that speaks of the use of molecular oxygen, doesn't it? A. I don't believe I can find that statement about molecular oxygen.

Q. Look in the equation, doesn't that show O_2? A. As far as the representative equation is concerned, if shows O_2.

Q. And O_2 is the chemical symbol for molecular oxygen? A. Certainly.

[729]

Q. Is it not true of both the original and Reissue patents that ethylene oxide is formed in the reaction zone by the direct combination of only ethylene and molecular oxygen in the presence of the catalyst and in the presence of water and at a temperature between 150 and 400 degrees centigrade? A. Will you repeat that question?

(Question repeated by the stenographer.)

By Mr. Watson:

Q. I would like a yes or no answer which you can then explain if you wish.

The Witness: That is a very long sentence. I wish you would repeat it once more.

(Question repeated by the stenographer.)

[730]

A. My answer is "Yes"; but the two patents differ in the requirement of what shall be present when the reaction occurs. The statement as read gives no evidence of what is happening on the catalyst surface, and that must be speculation.

Q. Well, we, of course, can not get inside the reaction zone, and we do not want to speculate about it. A. No, but that is the key to the thing.

Q. But as far as the process of operation procedure is concerned, your answer to that question was in the affirmative. A. Yes, the end product, one of the end products is the formation of ethylene oxide.

* * * * *

[733]

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Mr. Edmonds: I offer in evidence a copy of the article written by Dr. Reyerson and his associate Dr. Swearingon, published in the Journal of the American Chemical Society, in the Volume for July-December, 1928.

* * * * *

[1183]

* * * * *

Thereupon—

LLOYD H. REYERSON, a witness heretofore sworn, was recalled by the Defendant and examined and testified as follows:

CROSS-EXAMINATION BY MR. WATSON:

* * * * *

[1189]

* * * * *

Q. From the chemical viewpoint, isn't it true that the sine qua non of the process is that water shall always be present in the reaction zone? A. If the side reaction occurs water will always be present in the reaction zone.

Q. And so far as you know the side reaction will always occur? A. It usually does.

[1015]

Testimony of Walter Wilkinson.

Thereupon—

WALTER WILKINSON, a witness of lawful age, produced on behalf of the Defendant, in surrebuttal, having been first duly sworn, was examined and testified as follows:

* * * * *

[1026]

CROSS-EXAMINATION BY MR. WATSON:

Q. Are you a laboratory man? A. I have done work in the laboratory.

* * * * *

[1027]

A. I am a research man, and I am in the Research Department. I am not an operator.

* * * * *

[1314]

* * * * *

Testimony of Ray M. Carter.

Thereupon——

RAY M. CARTER, a witness recalled by the Defendant, testified further as follows:

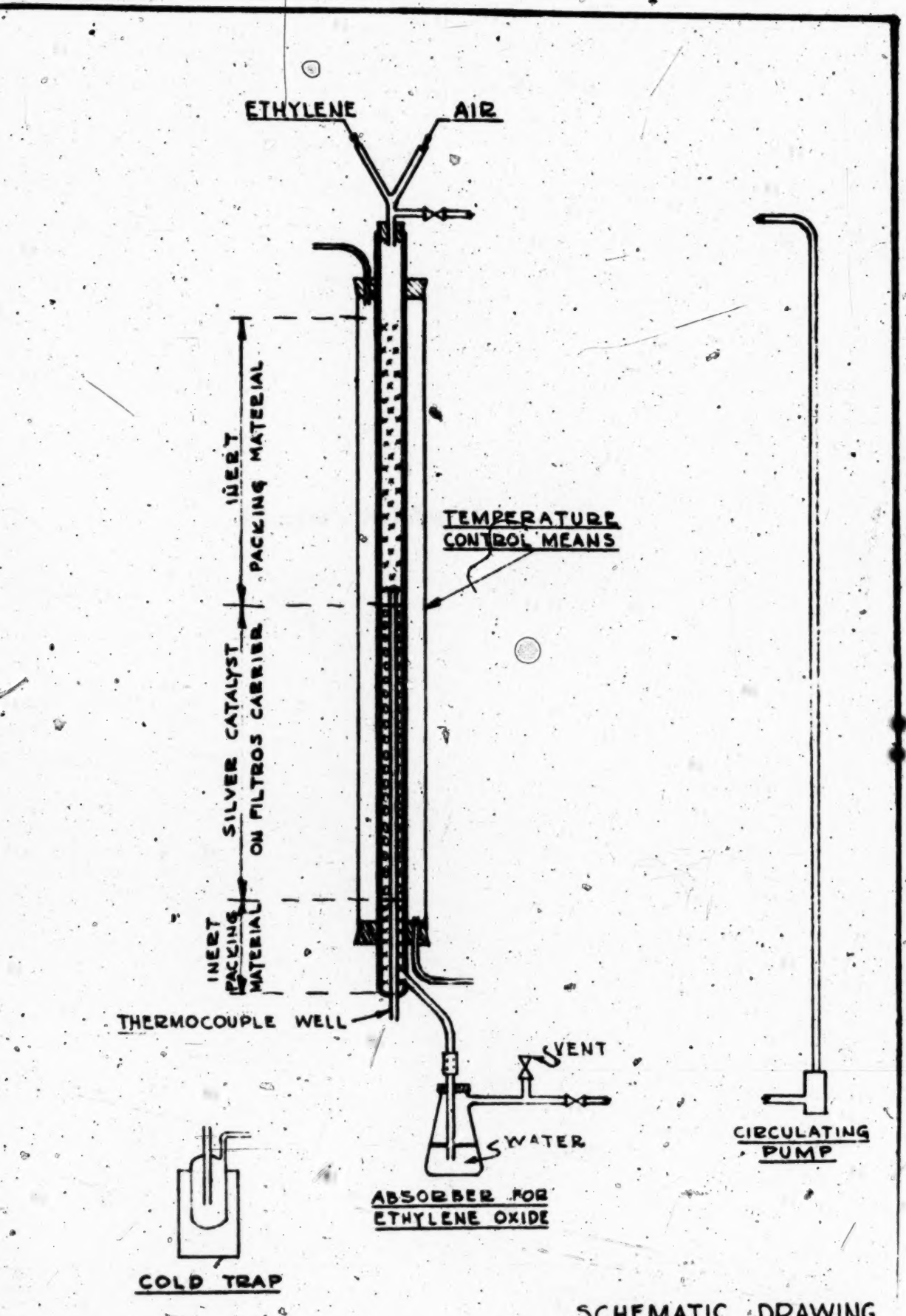
Mr. Barrows: In order to save time, and in accordance with the agreement last night, we wrote out the

[1315]

direct examination of the witness and furnished it to the other side last night. And it is stipulated that this may be written into the record as the direct examination, in lieu of taking the time to call the witness this morning.

The Court: Very well.

Plaintiff's Exhibit 4.



SCHEMATIC DRAWING
LEFORT PROCESS

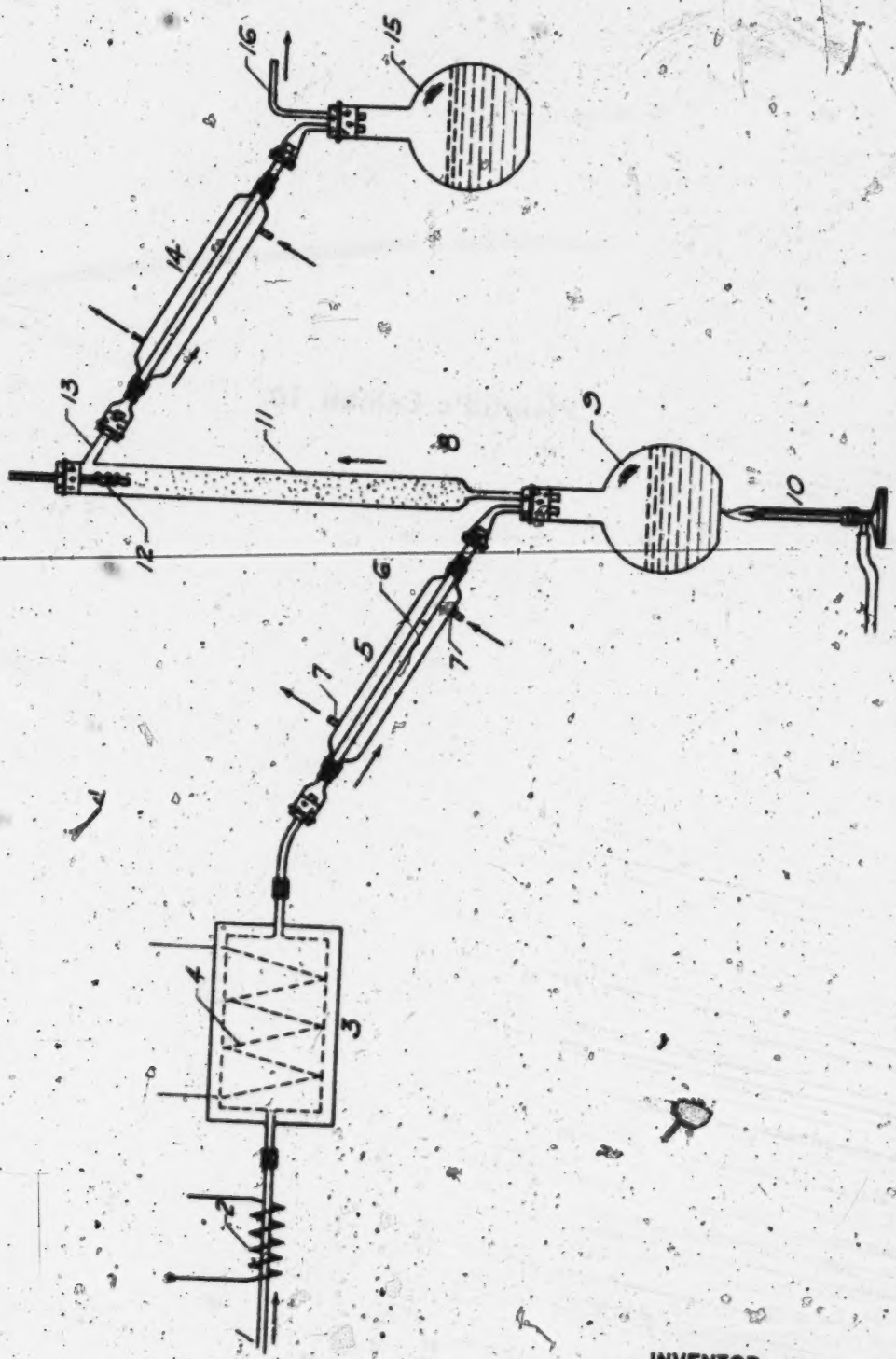
Plaintiff's Exhibit 10.

Plaintiff's Exhibit 10 (continued).

Aug. 8, 1939. H. S. TAYLOR 2,168,590

CATALYTIC ALKYLATION OF AROMATIC HYDROCARBONS

Filed Dec. 7, 1935



INVENTOR
Hugh S. Taylor
BY
Virgil F. Dawes
ATTORNEY

Plaintiff's Exhibit 10 (continued).

Patented Aug. 8, 1939 2,168,590

UNITED STATES PATENT OFFICE

2,168,590

CATALYTIC ALKYLATION OF AROMATIC HYDROCARBONS

Hugh S. Taylor, Princeton, N. J., assignor to The M. W. Kellogg Co., New York, N. Y., a corporation of Delaware

Application December 7, 1935, Serial No. 53,298

9 Claims. (Cl. 260—671)

This invention relates to catalytic processes of chemical synthesis and more particularly to processes involving the alkylation of organic compounds. More specifically, the invention relates to the production of valuable compounds by the direct reaction between less valuable materials composed preponderantly of the constituents of the desired compound under the influence of a suitable catalyst and under milder conditions than those employed in the prior art.

In the synthesis of many organic compounds the reactions involved are often of a very complex nature and the production of the desired product is achieved only by a multiple step-wise procedure usually involving the formation of numerous intermediate products and very often the formation of undesirable byproducts of little value resulting not only in a high manufacturing cost but also in a relatively low yield of the desired product. Moreover the operating conditions are sometimes of a drastic nature, often necessitating high temperatures, unusual pressures, or other features requiring costly equipment which may be subject to rapid deterioration due to one or more of the steps in the manufacturing procedure. As a result of these and other considerations it is a not unusual situation that the high cost of certain products precludes their general use for purposes for which they are eminently suited and where their employment, if not barred by the element of excessive cost, would be of great benefit to those so employing them.

The present invention has for one of its objects the provision of a simple and economic process for producing many desirable organic compounds which involves a minimum of procedural steps, employing relatively simple and inexpensive equipment, and requiring operating conditions considerably less drastic than those ordinarily necessary.

In the operation of my invention I accomplish the direct alkylation of many organic compounds, or in certain cases inorganic compounds, in a single step under unusually mild operating conditions with a relatively high yield of the desired product and with a minimum production of byproducts of lesser value. By the term alkylation is meant the substitution of radicals or alkyl groups for one or more of the hydrogen atoms or other atoms or radicals of a molecule, or it may mean the addition of such groups at the double bond or bonds, or triple bond or bonds, of a molecule, where such bond or bonds occur, without the displacement of any of the original components of the molecule.

To cause the desired reaction to proceed I choose a catalyst and operating condition such that I secure the activation of specific bonds in the reacting materials, the bonds being those which will, when activated, result in the desired reaction. This specificity of a catalytic surface will be made clear by the following very simple example made possible by the use of deuterium ("heavy hydrogen") whereby it is possible to note the exchange of hydrogen atoms in a molecule or, in effect, to label the atoms, a procedure obviously impossible before the discovery and isolation of this material. When ethane and deuterium were passed together over a given nickel catalyst at a temperature of 135° C. there occurred a replacement of hydrogen atoms in the ethane molecule by deuterium atoms, the replacement being substantially complete after an hour, the products being "heavy ethane" and hydrogen. This interchange of atoms would of course be undiscernible except for the use of deuterium. When hydrogen and ethane are similarly passed over the catalyst at this temperature the products are identical with the feed and there appears to have been no reaction, whereas in actuality there may have been a large scale interchange of atoms of hydrogen between the original ethane and the hydrogen. By the use of deuterium however, it is now possible to state definitely that such interchange has occurred thus showing positively the activation of the C—H bonds in the ethane. When, however, ethane and hydrogen were passed over the same catalyst at a temperature of 200° C. the dissociation of ethane occurred by the breakage of a C—C bond, resulting in the production of methane, thus,

$$C_2H_6+H_2 \xrightarrow{Ni} 2CH_4$$

This reaction was substantially complete, i. e., total conversion of C_2H_6 to CH_4, after about an hour. The reaction producing methane was first detectable at about 150° C. Thus it is quite evident that, with an active nickel catalyst, whereas at 135° C. the C—H bonds of ethane are activated, at 200° C. the C—C bonds are in addition activated, thus illustrating the specificity of the activation of the bonds by the catalyst. It appears almost needless to note that neither of these reactions can be detected at the temperatures used in the absence of the catalyst.

Other examples of the activation of certain bonds as detected by the use of deuterium are

Plaintiff's Exhibit 10 (continued).

the replacement of H atoms in benzene by deuterium at 200° C. over nickel, and over chromium oxide gel at somewhat higher temperatures and similarly the replacement of hydrogen atoms in CH_4 and NH_3 under conditions under which it was supposed that these materials were quite inert.

Having selected the catalyst and the operating conditions favorable to the activation of the desired bonds in the reactants involved it is now only necessary to contact the materials with the catalyst under the selected operating conditions and, after allowing the necessary reaction time to elapse, to withdraw the products, cooling, condensing, and otherwise handling these products as may be appropriate.

In order to make more clear the operation of my invention I will describe by way of illustration, but not of limitation, a specific embodiment of my invention whereby benzene and ethane are reacted to produce toluene according to the equation:

$$C_6H_6+C_2H_6 \xrightarrow{Ni} C_6H_5CH_3+CH_4$$

It will be observed that this reaction involves the alkylation of benzene by the substitution of a methyl group (CH_3) for one of the hydrogen atoms of the benzene, methane being produced as a byproduct in addition to toluene, the amount of this byproduct, however, being less than 15% by weight.

In the discussion of the specificity of catalytic surfaces, it was pointed out that at for example 200° C. active nickel activates C—C bonds of ethane and C—H bonds of benzene. It is readily seen that this catalyst and temperature may be employed to carry out this reaction. The process may be carried out at atmospheric pressure, subatmospheric, or superatmospheric pressure. For convenience and simplicity I prefer now to operate at substantially atmospheric pressure.

Referring now to the drawing, the single figure of which represents a form of apparatus suitable for carrying out the invention, a mixture of benzene and ethane is introduced at pipe 1 from a suitable container or compressor (not shown) and passes through a heating zone 2, diagrammatically shown. The heated mixture then passes into the insulated catalytic chamber 3 which contains the catalyst mounted preferably in pellet form on any suitable material as, for example, pumice or kieselguhr. The temperature of the catalyst chamber 3 may be maintained and controlled by means of the temperature regulating coil 4. The temperature and pressure at appropriate points in the apparatus may be determined by suitable instruments (not shown) installed wherever desired. The reacted product passes from the catalyst chamber through the cooler and condenser 5, furnished with a cooling medium jacket 6 having cooling medium entrance and exit 7. The greater portion of the condensible portion of the material passing through the condenser 5 is liquefied therein and the condensed material and uncondensed vapor and gas pass together into the fractionating equipment shown generally at 8. This equipment consists of condensate accumulator 9 which may be heated for reboiling by burner 10. Accumulator 9 may be provided with means (not shown) for periodically or continuously withdrawing condensate. Fractionating column 11 of any suitable design fractionates the vapor passing upwardly through it in the usual manner and may be furnished with reflux condensing coil 12 at its upper end and vapor outlet 13. The vapor passing through the outlet 13 passes to the condenser 14, similar to condenser 5. Condensate and uncondensible gases pass from the condenser 14 to the receiver 15 in which the condensate is accumulated, uncondensed gases passing out through tube 16 to an appropriate gas handling arrangement (not shown). The receiver 15 may be furnished with means (not shown) for withdrawing condensate as desired.

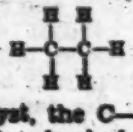
The raw materials, benzene and ethane, may be fed to the apparatus in any desired proportion, as for example in about equal molecular proportions or an excess of benzene may be used to insure the substantially complete conversion of the ethane, or, if desired, an excess of ethane may be used. Thus I may so proportion the feed that it contains two mols of benzene for each mol of ethane. The feed thus introduced is heated on its passage to the catalyst chamber to the desired degree, preferably in the neighborhood of 200° C. As noted hereinbefore the specificity of the particular catalyst cited, i. e., an active nickel, for the activation of the C—C bond of ethane was first noticeable at a temperature of about 150° C., therefore that temperature is the lowest at which the process is operable with such a catalyst. Also the process may be carried out at a temperature as high as 300° C. or higher but as already stated the preferred temperature is in the neighborhood of 200° C. It is to be understood that it is possible to carry out this reaction with different catalysts, those in the group classed as hydrogenation-dehydrogenation catalysts being generally suitable. None of these catalysts necessarily exhibits the same degree of catalytic effect at a given temperature as any other and for this reason different temperatures and rates of conversion would be used with a different catalyst. The specific operating conditions cited for the particular example described apply only for the catalyst cited and are not to be regarded as limiting conditions.

The free space within the catalyst chamber 3 is such that, for the given rate of feet, the reacting materials are maintained in contact with the catalyst for a period of time sufficient to allow the reaction to go forward to the desired extent. Under the conditions hereinbefore noted a reaction time of one hour, more or less, is satisfactory although, depending on conditions, a reaction time of two to five seconds or more may be sufficient while under other conditions a reaction of three hours or longer are desirable.

The material effluent from the catalyst chamber passing to the condenser 5 is composed of the desired product toluene and the byproduct methane in substantially equal molecular proportions along with any unconverted benzene and ethane. The liquefiable portions of this mixture, that is to say, the toluene and benzene, are largely condensed in condenser 5 although it is not essential that such liquefaction be entirely complete. The function of the fractionating apparatus 8 is to accomplish the separation of the product toluene from the more volatile benzene which has escaped conversion, as well as from any dissolved gaseous ethane and methane. The operation of such apparatus is well known and in this case toluene is recovered as a condensate in receiver 9 while the benzene and fixed gases pass overhead in vapor form through the outlet 13 to the condenser 14 where the benzene vapor is substantially completely liquefied, the liquid benzene being collected in the receiver 15. The

uncondensed material comprising the byproduct methane and any unconverted ethane are conducted away from the system by means of the tube 16 for such subsequent treatment as may be desired. The benzene collected in receiver 15 may be reintroduced into the feed to the apparatus following the customary procedure of recycling unreacted portions of a feed stock. As already noted it is preferable in this process so to proportion the feed that an excess of benzene is present in order to further the substantially complete conversion of the ethane since the recovery of unconverted ethane substantially free of methane is accomplished with difficulty. Depending on the thoroughness of the fractionation in this apparatus, the toluene may be recovered from receiver 9 as a substantially pure product or it may require subsequent purification by any suitable process.

As to the mechanism by which reactions of this type proceed, theoretical considerations and intensive study of catalytic phenomena in general, carried on over a considerable period of years lead me to believe at present that the reactions follow the course briefly outlined below. For the purpose of clarity the discussion will be confined, at the start at least, to the specific reactions of benzene and ethane. As already described, I have found that where ethane and hydrogen are passed over an active nickel at about 200° C., complete decomposition of the ethane to methane occurs. I believe that the ethane is chemically adsorbed on the surface of the catalyst in an activated condition in which the C—C bond is specifically activated. That is to say, when the ethane molecule

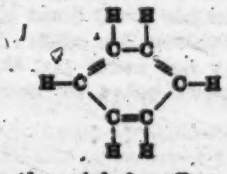


contacts the catalyst, the C—C bond is broken and the material is adsorbed on the surface as two methyl (CH_3) radicals, thus:

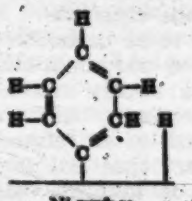
It may be considered that a methyl nickel complex is formed. Similarly hydrogen (H_2) on contacting the nickel surface is adsorbed in an activated state as two atoms of hydrogen. When ethane and hydrogen in mixture are thus adsorbed, we may picture the adjacent adsorbed fragments thus:

Under the conditions imposed there is a continual interaction between adjacent adsorbed radicals; thus it becomes apparent that adjacent CH_3 and H groups may readily unite to form relatively stable methane (CH_4) molecules which would thus appear as a product. It is also apparent that interaction between adjacent CH_3 groups may occur, but this merely regenerates C_2H_6 and interaction between adjacent activated H atoms merely regenerates H_2.

With regard to the activated adsorption of benzene on the same catalyst under like conditions, it has already been pointed out that in this case it is the C—H bonds of the benzene molecule that are activated, as shown by the experiments with deuterium. This type of activated adsorption may be pictured thus. The benzene molecule,

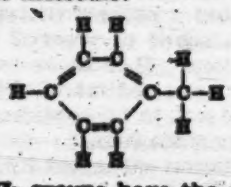


on contacting the nickel suffers a rupture of a C—H bond whereby a C_6H_5 radical and an H atom are adsorbed on the catalytic surface:



This type of adsorption is demonstrated by the fact that when deuterium is present, deuterium is found to be substituted for H in the molecule.

This rather detailed discussion is included by way of substantiating my theory as to the mechanism of the ethane-benzene reaction. In the light of the above it becomes rather simple to account for the production of toluene and methane from benzene and ethane under the influence of the catalyst. The ethane is adsorbed in an activated condition as CH_3 radicals and the benzene is adsorbed as C_6H_5 radicals and H atoms. Thus adjacent CH_3 and C_6H_5 groups will occur with a frequency dependent on the relative concentrations of the two materials on the surface, and reactions therebetween are to be expected, forming the toluene molecule:

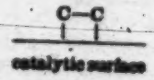


Also H and CH_3 groups have the opportunity to react to form methane and free energy calculations indicate that the joint occurrence of these two reactions is favored. The formation of diphenyl by the interaction of two adjacent C_6H_5 groups with simultaneous formation of H_2 is not favored by the free energy relationships involved, nor is the formation of ethyl benzene, and the presence of these products has not been noted in the end products of the reaction. Interaction between adjacent CH_3 groups will, of course, merely regenerate ethane and interaction between adjacent C_6H_5 and H groups results in the regeneration of benzene.

This reaction as above described is illustrative of the type of reaction broadly included within the scope of my invention and is characterized by the activation of specific bonds at catalytic surfaces and the reaction between the thus activated residues derived from distinct compounds to form specific desired products. The reaction detailed above is one wherein a C—H bond in one of the reactants is activated and a C—C bond in the other is activated and the residues thus activated interact to produce the desired product. Depending on the materials used, the catalyst, and conditions employed, any specific type of bond in one material may be activated and another specific bond in the other may be acti-

Plaintiff's Exhibit 10 (continued).

vated, that is to say, the bonds involved may be C—C, C—H, C—N, C—O, O—H, N—H, etc. In the case of the double C=C bond and the triple C≡C bond the situation is modified in that complete rupture may not occur but the bond may be activated as shown below for double bond:



so that a product is formed by the addition of an activated residue at each of the free bonds, i. e., those bound to the catalyst.

It is of course to be understood that products cannot be formed indiscriminately without regard to the free energy relationships involved and that it is necessary that the free energy of the products must be so related to that of the reacting materials that the production of the desired product is favored. Because of these free energy relationships, the temperature required for purely thermal activation of the reacting materials is, in a great many cases, of such magnitude that the free energy change of the desired reactions is unfavorable, whereas it is possible to carry on such reactions utilizing the specific activation by the catalytic surface at a lower temperature, where the free energy change of the reaction favors its completion. Thus it is clear that the specific activating properties of the catalyst make possible the carrying out of many desirable reactions, which could not otherwise be conducted at any temperature in the absence of the catalyst, whereby the usual conception of the action of a catalyst in merely lowering the reaction temperature is modified to include making possible reactions which could not otherwise be made to proceed.

Below are listed a number of typical reactions which may be caused to proceed in accordance with my invention. It is to be understood that these examples are cited merely by way of illustration and are not to be considered as limiting the scope of my invention.

Reactions between saturated hydrocarbons:

$$C_{n_1}H_{2n_1+2}+C_{n_2}H_{2n_2+2}=2\frac{C_{n_1+n_2}H_{2n_1+2n_2+2}}{2}$$

or

$$=C_{n_1+n_2-1}H_{2(n_1+n_2)}+CH_4$$

Reactions between paraffins as shown above by these general equations may be caused to proceed under proper conditions in the presence of a suitable catalyst as described. Obviously combinations of these reactions may occur when conditions favor each of them. These reactions illustrate generally the type of reactions possible between aliphatic hydrocarbons of which, of course, there are numerous variations possible, as for example, when unsaturated hydrocarbons are used.

Reactions between aromatics and saturated aliphatics are exemplified below:

$$C_6H_6+C_nH_{2n+2}=C_6H_{6-y}(CH_3)y+C_{n-y}H_{2(n-y)+2}$$

where

$$x+y=6$$

and

$$n>y$$

For example:

$$C_6H_6+C_2H_6=C_6H_5CH_3+CH_4$$

and

$$C_6H_6+C_2H_6=C_6H_4(CH_3)_2+CH_4$$

Here again numerous variations and combinations are possible some of which result in the formation of multi-ring compounds and may be exemplified by the general reaction:

$$nC_6H_6R'+mC_6H_6R''=(C_6H_4)_{n+m}+R'_nR''_m$$

Reactions between paraffins and ammonia to form amines and H_2 are not favored thermodynamically but those forming amines and a paraffin hydrocarbon are favored and may be generally shown thus:

$$RR_1+NH_3=RNH_2+RH$$

Those between ammonia and olefines can be carried out successfully, particularly when the number of carbon atoms in the hydrocarbon is relatively small. The general reaction may be expressed as below:

$$C_nH_{2n}+NH_3=C_nH_{2n+1}NH_2$$

Similarly OH groups, NH_2, CO, CHO, halide, nitrogen, and sulphur groups and the like may be substituted in various hydrocarbons, their derivatives, and other organic compounds.

In general the catalysts employed in the operation of my invention are those generally classed as hydrogenating-dehydrogenating catalysts of which a great many are known such as for example, nickel, palladium, platinum, copper, iron, oxides of nickel, chromium, copper, zinc, and many other metals, their oxides, sulphides and other compounds thereof and mixtures of the same. It must not be inferred from this that the action of these various materials is identical and that any one of them may be used for any reaction. The extent to which they catalyze the various reactions differs markedly from one material to another and their specificity in the activation of certain bonds is also different. However, by selecting that catalyst which is found to activate the desired bonds under the proper conditions, these reactions may be caused to proceed in the direction of the desired products.

I claim:

1. The process of alkylating aromatic hydrocarbon compounds by paraffinic hydrocarbon compounds which comprises activating a bond external to the aromatic ring of an aromatic hydrocarbon compound by subjecting it to the action of a hydrogenating-dehydrogenating catalyst having specificity properties for said external bond, under such conditions of temperature and pressure that said catalyst specifically activates said external bond, simultaneously activating a carbon to carbon bond of a paraffinic hydrocarbon compound by the action of said catalyst in the presence of said activated aromatic hydrocarbon compound to form activated residues of said paraffinic hydrocarbon compound whereby said activated residues react with and alkylate said activated aromatic hydrocarbon compound.

2. The process of alkylating aromatic hydrocarbon compounds by paraffinic hydrocarbon compounds which comprises activating a bond external to the aromatic ring of an aromatic hydrocarbon compound by subjecting it to the action of a hydrogenating-dehydrogenating catalyst having specificity properties for said external bond, under such conditions of temperature and pressure that said catalyst specifically activates said external bond, simultaneously activating a bond of a paraffinic hydrocarbon compound by the action of said catalyst in the presence of said activated aromatic hydrocarbon compound to form activated residues of said paraffinic hydrocarbon compound whereby said activated residues react with and alkylate said activated aromatic hydrocarbon compound.

Plaintiff's Exhibit 10 (continued).

3. The process of alkylating aromatic hydrocarbon compounds by paraffinic hydrocarbon compounds which comprises activating bonds external to the aromatic ring of an aromatic hydrocarbon compound by subjecting it to the action of a hydrogenating-dehydrogenating catalyst having specificity properties for said external bond, under such conditions of temperature and pressure that said catalyst specifically activates said external bonds, simultaneously activating a carbon to carbon bond of a paraffinic hydrocarbon compound by the action of said catalyst in the presence of said activated aromatic hydrocarbon compound to form activated residues of said paraffinic hydrocarbon compound whereby said activated residues react with and alkylate said activated aromatic hydrocarbon compound.

4. The process of methylating aromatic hydrocarbon compounds which comprises subjecting ethane to the action of a hydrogenating-dehydrogenating catalyst under conditions such that said catalyst having specificity properties for the carbon to carbon bond of ethane, specifically activates the carbon to carbon bond of the ethane to form activated methyl groups, simultaneously subjecting an aromatic hydrocarbon compound to the action of said catalyst in the presence of the ethane, said catalyst and said conditions being also such that said catalyst specifically activates a bond external to the aromatic ring of said aromatic hydrocarbon compound, whereby said activated methyl groups react with and methylate said activated aromatic hydrocarbon compound.

5. The process of synthesizing alkylated benzenes which comprises subjecting benzene and saturated paraffin hydrocarbon of the general formula C_nH_{2n+2}, where n is 2 or greater, to the action of a hydrogenating-dehydrogenating catalyst at a temperature and for an interval of time sufficient to produce a substantial yield of alkylated benzene.

6. The process of synthesizing toluene from benzene and ethane which comprises subjecting ethane to the action of a hydrogenating-dehydrogenating catalyst having specificity properties for the carbon to carbon bond of ethane, under conditions of temperature and pressure such that said catalyst specifically activates the carbon to carbon bond of the ethane to form activated methyl groups, simultaneously subjecting benzene to the action of said catalyst in the presence of the ethane, said catalyst and said conditions being also such that said catalyst specifically activates a bond external to the aromatic ring of said benzene, whereby said activated methyl groups react with and methylate said activated benzene to form toluene.

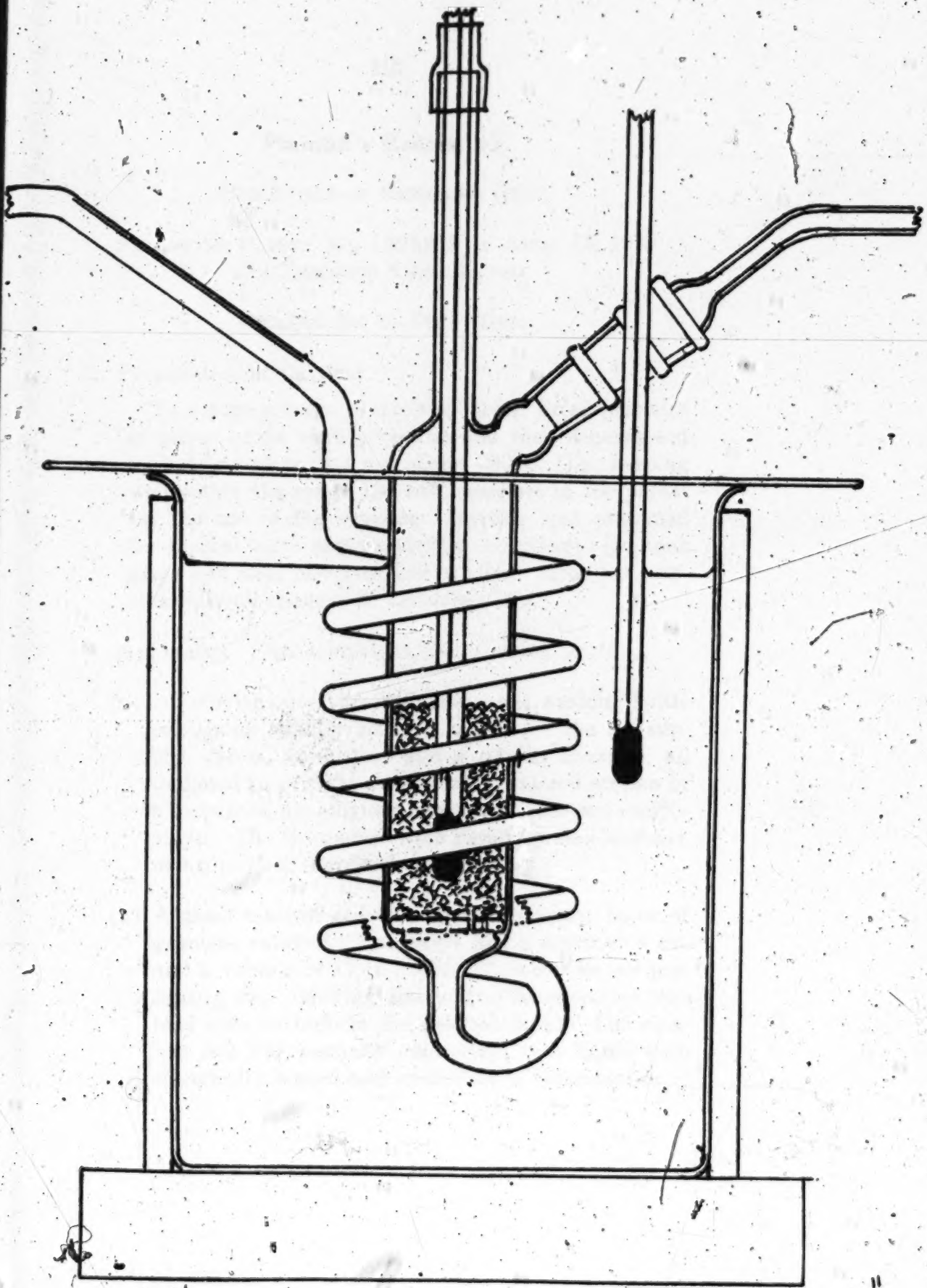
7. The process of synthesizing toluene from benzene and ethane which comprises subjecting ethane to the action of a hydrogenating-dehydrogenating catalyst having specificity properties for the carbon to carbon bond of ethane under conditions of temperature and pressure such that said catalyst specifically activates the carbon to carbon bond of the ethane to form activated methyl groups, simultaneously subjecting benzene to the action of said catalyst in the presence of the ethane, said catalyst and said conditions being also such that said catalyst specifically activates an external bond of said benzene, whereby said activated methyl groups react with and methylate said activated benzene to form toluene.

8. The process of synthesizing toluene from benzene and ethane which comprises subjecting ethane to the action of an active nickel catalyst under conditions such that said catalyst specifically activates the carbon to carbon bond of the ethane to form activated methyl groups, simultaneously subjecting benzene to the action of said catalyst in the presence of the ethane, said catalyst and said conditions being also such that said catalyst specifically activates an external bond of said benzene, whereby said activated methyl groups react with and methylate said activated benzene to form toluene.

9. The process of synthesizing toluene from benzene and ethane which comprises subjecting benzene and ethane to the action of an active nickel catalyst at a temperature ranging from 150° C. to 300° C. for a time interval sufficient to produce a substantial yield of toluene.

HUGH S. TAYLOR.

Plaintiff's Exhibit 11.



Plaintiff's Exhibit 12.

PRODUCTION OF ETHYLENE OXIDE

FOLLOWING PATENT No. 1,998,878 OF APRIL 23, 1935
TO THEODORE EMILE LEFORT

Catalyst No. 1—Pure Silver

1. Preparation of Catalyst

To secure a large surface of silver, dried granules of silver oxide were prepared and then decomposed by heating up to, but not above, 390°C. (i.e., keeping well within the range 150-400° specified in the patent for the use of the catalyst. Heating was continued for several hours until no further evolution of gas took place and until the dark brown silver oxide had substantially all changed to metallic silver.

2. Apparatus. This comprised the following parts:

(a) An ethylene pressure cylinder, a gas washing bottle containing slightly acidified water for the air supply, valves, flowmeters and a mixing chamber, all designed to provide a constant, regulated stream of a moistened air-ethylene mixture of constant composition. The flowmeters were carefully standardized, using air and ethylene, respectively.

(b) A glass catalyst cell to hold the columnar layer of granular catalyst. This layer had a depth of 4 cm. and a volume of 17 cc. The cell had coils for preheating the inflowing gas and a thermometer was held with its bulb in the catalyst layer. The catalyst cell was partially submerged in a liquid bath electrically heated and containing a thermometer.

Plaintiff's Exhibit 12 (continued).

(c) A train of Leidner wash bottles containing solutions which would absorb any ethylene oxide or carbon dioxide present in the gas issuing from the catalyst cell.

3. Procedure

Air was introduced at a rate of very nearly 500 cc. per minute; ethylene in most cases at 10 cc. but in one at 5 cc. per minute. This gave a mixture of approximately two (or one) per cent of ethylene in air which is just below the recognized lower explosive limit of such mixtures.

The temperature of the bath and the catalyst could be raised gradually by increasing the electric current in the heater under the bath. Convection currents in the bath liquid were rapid enough so that, with air alone flowing through the cell, there was only a slight differential between the reading of the thermometer in the catalyst and that in the bath. In the first run the solution for collecting and detecting the ethylene oxide was a hot, acidified, saturated salt solution containing methyl red as an indicator whose change in color showed the complete consumption of the hydrochloric acid by the ethylene oxide which dissolves. In later runs a saturated solution of sodium chloride containing a known amount of hydrochloric acid and also various indicators, was used to absorb the ethylene oxide. The carbon dioxide was partially collected by bubbling the gas issuing from the ethylene oxide absorbers through wash bottles containing known amounts of standardized sodium hydroxide solution. In some final runs a solution of nearly saturated magnesium chloride in water was used to collect the ethylene oxide as this gave a more complete absorption than the sodium chloride solution. In one of these runs the

Plaintiff's Exhibit 12 (continued).

carbon dioxide was also collected quantitatively by passing the gas mixture leaving the last ethylene oxide absorber through two Emmerling towers in series charged with concentrated sulphuric acid and then to two weighed U tubes filled with potassium hydroxide pellets.

To carry out a run, the controlled mixture of ethylene and air was passed through the catalyst at a given temperature until a steady state had been established as indicated by the readings of the thermometers. The effluent gas from the catalyst cell was then by means of a two-way stopcock made to pass through the absorbers, frequent readings of the flowmeters and the thermometers being made. After a suitable interval, as measured by a stopwatch, the flow through the absorbers was discontinued. The amount of ethylene oxide in the absorbers was then determined by titration. Similarly, the sodium hydroxide solution used for absorbing the carbon dioxide was titrated and the amount of carbon dioxide absorbed thus ascertained, or the U tubes were re-weighed.

4. Results

(a) In the first run, using a gas stream containing 500 cc. of air and 10 cc. of ethylene and slowly raising the temperature of the bath, a slight rise in the temperature of the catalyst as compared with that of the bath liquid was observed when the catalyst temperature reached 190°C., the differential increasing as the temperature rose. At the same time the warmed, saturated salt solution to which about ½ cm. of tenth molar hydrochloric acid and some methyl red had been added, through which the effluent gases bubbled, changed to a yellow color in about 3 minutes. Similar tests with repeated additions

Plaintiff's Exhibit 12 (continued).

of small amounts of tenth molar hydrochloric acid up to 6 cc. and with various indicators (methyl red, brom thymol blue and phenolphthalein) showed complete consumption of the acid by the ethylene oxide.

(b) In a second run the same mixture was passed through the catalyst at an average constant temperature of 213.5°±1 for 15 minutes and then through two wash bottles at room temperature containing an excess of the same absorbing solution. Analysis showed 28.7% conversion of the ethylene into ethylene oxide.

(c) In a third run an additional wash bottle containing a measured volume of standardized sodium hydroxide solution was attached to the end of the train to collect carbon dioxide in the effluent gas from the catalyst. To guard against any carbon dioxide in the inlet gas, a wash bottle containing sodium hydroxide solution was also introduced into the air supply ahead of the wash bottle containing dilute acid. Collecting the gas over a 15 minute interval, at an average catalyst temperature of 232°, 33.6% of the ethylene was converted to ethylene oxide and 6.3% to carbon dioxide. A repetition of this run at an average catalyst temperature of 235.5° gave a conversion of ethylene to ethylene oxide of 27.1% and to carbon dioxide of 6.1%.

(d) In subsequent runs, to guard against a possible slight loss of ethylene oxide from the second wash bottle, an additional (third) wash bottle was added containing an additional amount of the same absorbing solution. Several runs were made under otherwise identical conditions at catalyst temperatures of 215.5°, 229.0° and 244.7°. These gave an increased conversion both to ethylene oxide and to carbon dioxide as the temperature increased.

Plaintiff's Exhibit 12 (continued).

(e) A single run was made with a poorer mixture of ethylene, namely, 505 cc. of air and 5.05 cc. of ethylene per minute at a catalyst temperature of 218°. Conversions of 37.4% of the ethylene to ethylene oxide and 9.1% to carbon dioxide were obtained. These conversions were higher than would have been obtained at the same temperature in the richer gas (2% ethylene).

(f) Three runs were made using magnesium chloride solutions in the absorbers for the ethylene oxide, 515—520 cc. of air and 10.0 cc. of ethylene per minute at catalyst temperatures of 227.1, 227.5 and 230.3°C. Conversions of 34.1, 34.0 and 37.7% respectively of the ethylene to ethylene oxide were obtained.

In the final run the carbon dioxide was determined quantitatively and a conversion of 43.7% of the ethylene into carbon dioxide was obtained.

The data for the several runs are collected in the following table and the percentage conversions have been plotted in the accompanying diagram.

SPACE VELOCITY PER HOUR=1835±1%

No.	Flow cc./min. Air	Flow cc./min. C_2H_4	Temperature C° Catalyst	Temperature C° Bath	Bottles for C_2H_4O	Collection Interval min.	Conversion Vol. % C_2H_4O	Date
1	515	10.2	234	213.6	2	15	28.7	May 26
2	510	9.9$_5$	232	212.0	2	15	33.6	" 26
3	510	10.0$_0$	235.5	214.5	2	15	27.1	" 29
4	505	9.9$_5$	230.2	212.0	3	15	34.9	" 31
5	505	5.0$_5$	230.2	218.3	3	30	37.4	" 31
6	505	10.0$_5$	215.5	202.1	3	20	26.5	June 1
7	510	9.9$_5$	244.7	223.3	3	15	37.5	" 1
8	510	9.9$_5$	229.0	212.0	3	15	33.5	" 1
9	515	10.0$_0$	227.1	209.5	3	15	34.1	" 9
10	520	10.0$_0$	227.5	209.6	3	15	34.0	" 9
11	515	10.0$_0$	230.3	211.5	3	15	37.7	" 9

Plaintiff's Exhibit 12 (continued).

Catalyst No. 2—Silver on Earthenware

1. Preparation of Catalyst.

To secure, if possible, a large surface of silver from a relatively small weight of the metal, we impregnated 50 cc. of granular, unglazed earthenware by boiling it with a nearly saturated solution of silver nitrate for about an hour. After draining off the excess solution the material was dried overnight at 115°C. The dried granules were then heated until no more nitrate fumes were given off. This required prolonged heating overnight at temperatures of 440-463°C. Examined under the microscope the bright silver particles could be seen over the earthenware base, but the silver was more lustrous than that of Catalyst No. 1.

2. Apparatus and Procedure

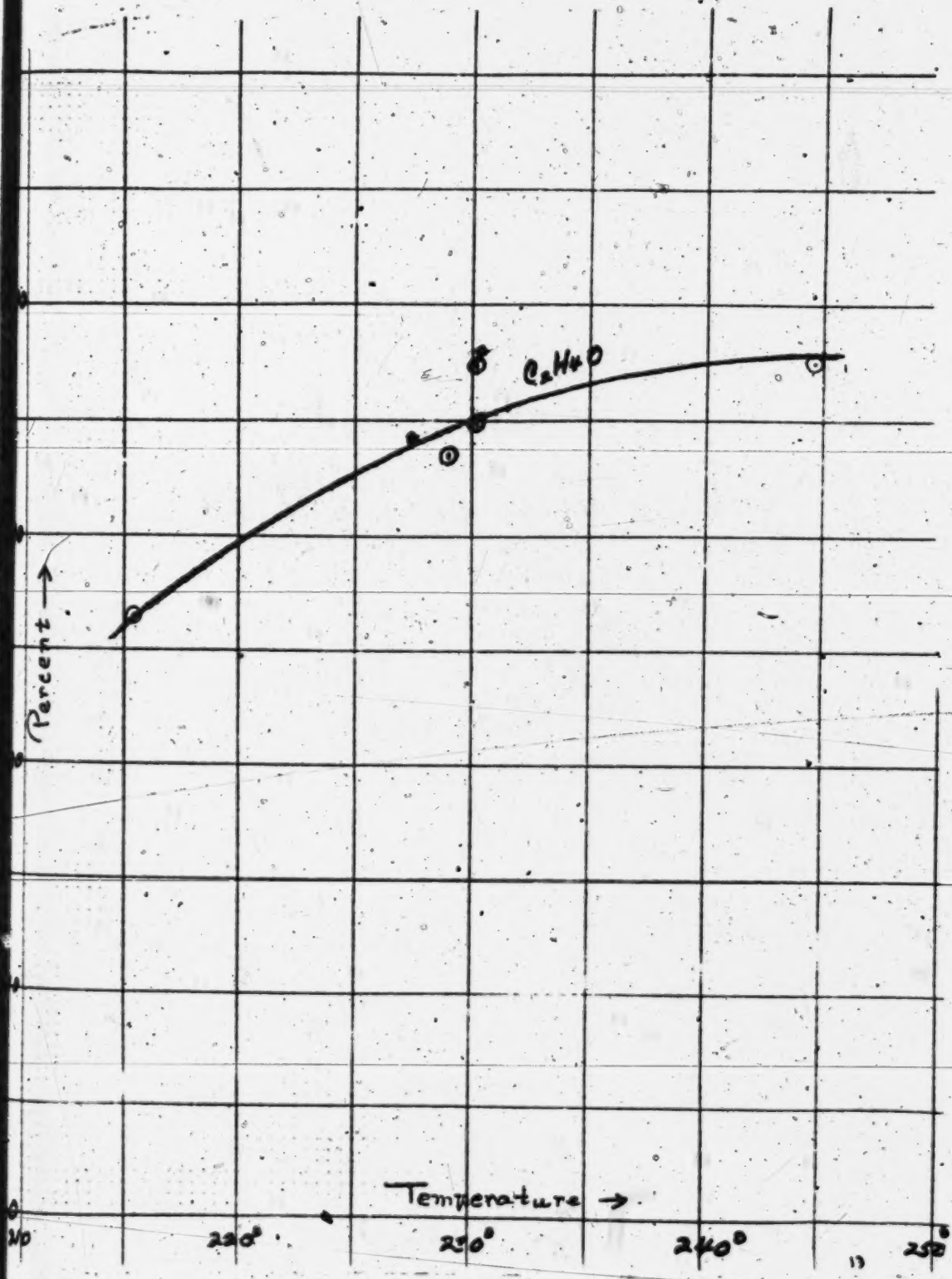
These were as before except that a final collection period of 30 minutes was employed.

3. Results

The catalyst temperature was slightly higher (3-4°) than the bath temperature at a temperature of 190°, but the differential increased but slightly as higher temperatures were reached (4-5° at 245°C.). The sample of effluent gas collected at this latter temperature (245°) on analysis showed only 0.9% conversion of ethylene to ethylene oxide and 0.24% to carbon dioxide.

June 3, 1940

Plaintiff's Exhibit 13.



Plaintiff's Exhibit 19.

AVERAGE DAILY WATER VAPOR CONTENT IN AIR CALCULATED AS PER CENT BY VOLUME FOR BALTIMORE, MARYLAND

APRIL 1st, 1939 — APRIL 30th, 1940

	1939									1940			
	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	Apr.
1	0.95	0.53	1.775	1.995	2.28	2.04	1.455	0.715	0.70	0.18	0.34	0.56	0.615
2	0.52	0.53	1.265	1.65	2.425	2.235	1.035	0.565	1.015	0.22	0.345	0.56	0.535
3	0.45	0.54	1.425	1.575	2.665	2.265	1.05	0.56	0.645	0.18	0.31	0.76	0.81
4	0.33	0.60	1.605	2.385	2.34	2.295	1.27	0.525	0.555	0.235	0.36	0.69	1.11
5	0.48	0.80	1.21	2.345	1.795	1.65	1.415	0.82	0.535	0.215	0.515	0.585	0.565
6	1.05	1.13	1.48	2.31	1.875	1.45	1.81	0.63	0.70	0.24	0.645	0.48	0.495
7	0.36	1.47	1.23	2.71	2.25	2.11	1.63	0.685	0.72	0.23	0.575	0.44	0.77
8	0.47	1.61	1.94	2.94	2.665	2.355	1.685	0.67	0.40	0.355	0.445	0.49	1.20
9	0.46	2.02	2.08	2.615	2.815	2.03	2.17	0.535	0.50	0.215	0.55	0.45	0.845
10	0.78	1.03	2.04	2.29	1.575	2.16	2.04	0.675	0.73	0.245	0.715	0.335	0.70
11	1.24	0.89	2.415	1.29	1.95	1.26	1.37	0.55	0.54	0.335	0.65	0.255	0.88
12	0.47	0.55	1.48	1.715	2.20	1.315	1.24	0.565	0.32	0.575	0.59	0.22	0.96
13	0.35	0.71	1.875	2.055	2.625	1.625	0.925	0.52	0.56	0.635	0.66	0.315	0.31
14	0.51	0.77	1.735	2.115	2.815	1.625	0.715	0.585	0.49	0.89	0.60	0.825	0.485
15	1.01	0.78	1.88	1.23	2.80	1.885	0.535	0.535	0.39	0.405	0.24	0.495	1.165
16	0.69	0.89	2.505	1.135	2.70	2.26	0.755	0.635	0.53	0.435	0.24	0.45	1.165
17	0.80	1.19	2.60	1.69	2.625	1.655	0.73	0.715	0.65	0.23	0.395	0.53	1.235
18	1.24	1.30	1.495	1.945	2.725	0.985	0.53	0.86	0.72	0.205	0.575	0.695	1.325
19	1.10	1.475	1.91	1.92	2.645	1.41	1.135	1.01	0.66	0.125	0.795	0.66	0.975
20	0.68	1.80	2.39	1.915	2.695	1.775	1.43	0.475	0.87	0.165	0.675	0.74	0.95
21	0.94	1.69	1.485	1.755	2.57	1.585	1.445	0.495	0.38	0.22	0.35	0.28	0.75
22	0.73	2.095	1.715	1.74	1.705	1.29	1.135	0.565	0.35	0.27	0.335	0.445	0.505
23	0.66	2.17	1.945	2.105	2.13	1.70	0.605	0.575	0.31	0.275	0.345	0.16	0.615
24	0.90	1.08	1.555	2.54	2.47	1.72	0.805	0.51	0.21	0.325	0.455	0.16	0.705
25	1.17	1.35	1.635	2.52	2.36	2.045	1.05	0.485	0.31	0.24	0.375	0.165	0.885
26	1.70	1.77	1.465	2.66	2.175	1.745	1.61	0.27	0.43	0.125	0.18	0.295	0.665
27	1.54	2.17	1.91	2.71	1.955	1.695	1.825	0.355	0.32	0.155	0.43	0.56	0.565
28	0.99	2.24	2.02	2.655	1.905	1.725	1.22	0.53	0.25	0.18	0.55	0.655	0.64
29	0.84	1.935	2.495	2.42	1.77	2.185	0.57	0.48	0.49	0.23	0.445	0.895	0.85
30	0.63	1.93	2.54	2.66	1.785	2.27	0.885	0.665	0.36	0.265		1.295	1.11
31		1.79		2.265	1.92		1.065		0.32	0.285		1.005	
Mo. Ave.	0.80	1.317	1.837	2.124	2.297	1.802	1.198	0.592	0.515	0.278	0.473	0.528	0.799
Cumul. Mo. Ave.	0.80	1.06	1.186	1.5195	1.675	1.696	1.625	1.496	1.387	1.276	1.203	1.147	1.12
Yearly Ave.													

	Reissue No. 20,370		No. 1,998,878	
	Application September 25, 1936	Issued May 18, 1937	Application March 22, 1932	Issued April 23, 1935

LEFORT ORIGINATED A PROCESS IN WHICH ETHYLENE OXIDE IS OBTAINED DIRECTLY FROM ETHYLENE AND MOLECULAR OXYGEN BY THE DIRECT ADDITION OF ONE ATOM OF OXYGEN TO ONE MOLECULE OF ETHYLENE.

P. 1, col. 1, lines 1-4
"This invention provides a specific and novel process for making ethylene oxide. It essentially consists in causing ethylene to combine directly with molecular oxygen . . ."

P. 1, col. 1, lines 12-14
"This process is characterized by an entirely unique reaction in which the direct addition of a single oxygen atom to the ethylene molecule. . ."

P. 1, col. 1, lines 17-20
"This novel reaction may be expressed as:

$$2CH_2{=}CH_2 + O_2 \longrightarrow 2CH_2{-}CH_2"$$
(O)

P. 1, col. 1, lines 1-2
"This invention has for object a process for the production of ethylene oxide . . ."

P. 1, col. 1, lines 9-14
". . . the reaction which can be written as follows:

$$2\ \begin{matrix}H_2C\\ \|\\ H_2C\end{matrix} + O^2 \longrightarrow 2\ \begin{matrix}H_2C\\ \diagdown O \diagup\\ H_2C\end{matrix}"$$

LEFORT DISCOVERED THAT ETHYLENE OXIDE IS THUS PRODUCED BY SUBJECTING A MIXTURE CONTAINING ETHYLENE AND MOLECULAR OXYGEN TO HIS COMBINATION OF SILVER SURFACE CATALYST AND CONTROLLED TEMPERATURE BETWEEN 150° AND 400° C., WITH WATER ALWAYS PRESENT IN THE REACTION ZONE.

P. 1, col. 1, lines 2-8
"It essentially consists in causing ethylene to combine directly with molecular oxygen at temperatures of about 150° to about 400° C. in the presence of a surface catalyst which favors the oxidation of ethylene to ethylene oxide under these conditions."

P. 1, col. 1, lines 26-36
"Ethylene from any desired source may be used, including that obtained from coke oven gas, hydrogenation of acetylene, or dehydration of ethyl alcohol. The oxygen can be the oxygen of the air, and can conveniently be mixed with the ethylene prior to the reaction.
"The oxidation of ethylene takes place with a giving off of heat, and it is, of course, desirable to maintain the temperature of the zone of reaction within the range specified."

P. 1, col. 1, lines 43-54
"The process can be conveniently conducted by passing a mixture containing air and ethylene through a tube, for example, of silica or of a material capable of withstanding high pressures, within which is placed such a surface catalyst as one composed of silver in finely divided form, and which may be disposed within the tube on a supporting material, or in other known ways. The temperature of the reaction zone is maintained between about 150° and about 400° C., for example, at about 200° C., and the issuing products from the tube contain ethylene oxide, . . ."

P. 1, col. 2, lines 12-17
"In any case, the ethylene and oxygen are thus reacted simultaneously at the temperatures set forth in the presence of a surface catalyst and of water, and the process is conducted to produce ethylene oxide as a direct product in substantial and economic amount."

P. 1, col. 1, lines 1-6
"This invention has for object a process for the production of ethylene oxide which mainly consists in subjecting ethylene to the simultaneous action of the oxygen of air and of water, in presence of a catalyzer and, if need be, of hydrogen."

P. 1, col. 1, lines 16 and 20-21
"This reaction takes place . . . at a temperature between 150 and 400° C."

P. 1, col. 1, lines 22 and 26
"The following catalyzers may be used:
"(2) Silver alone."

P. 1, col. 2, lines 6-9
"The ethylene can be obtained from any source of supply: gas from coke ovens, hydrogenation of acetylene, dehydration of ethyl alcohol, and the like."

P. 1, col. 2, lines 12-13
"The oxygen can be the oxygen of the air, this latter gas being introduced in the reaction."

P. 2, col. 1, lines 28-33
"Silver in powder form, or in the form of a wire gauze, and placed or not on a support or carrier, is introduced in a tube, made for instance of silica, and, after the said tube has been heated to a temperature comprised between 150 and 400° C. . . ."

IN LEFORT'S PROCESS, THE ETHYLENE OXIDE IS PRODUCED WITHOUT SUBSTANTIAL FORMATION OF ALDEHYDES BUT WITH BURNING OF SOME ETHYLENE TO WATER AND CARBON DIOXIDE.

P. 1, col. 1, lines 15-25
". . . without the formation of significant amounts of other oxygenated compounds such as aldehydes. This novel reaction may be expressed as:

$$2CH_2{=}CH_2 + O_2 \longrightarrow 2CH_2{-}CH_2$$
O

and it is accompanied by complete oxidation of some of the ethylene to give carbon dioxide and water according to the equation:

$$CH_2{=}CH_2 + 3O_2 \longrightarrow 2CO_2 + 2H_2O"$$

P. 1, col. 1, lines 53-55
". . . the issuing products from the tube contain ethylene oxide, carbon dioxide and water without objectionable aldehydes."

P. 1, col. 2, lines 14-18
"The efficiency of the reaction is increased by diminishing the CO_2 which is formed, by introduction, in this reaction, of a suitable quantity of water. A suitable volume of CO_2 can also be previously introduced in the reacting gases."

P. 1, col. 2, lines 29-35
"Aldehydes formed exist only as traces.
"Moreover, the applicant has found that the reaction giving CO_2 is, contrarily to previous belief, a reaction of oxidation independent from that giving ethylene oxide and from that giving aldehydes."

P. 1, col. 2, line 41

$$"C_2H_4 + 3O_2 = 2H_2O + 2CO_2"$$

French Patent No. 739,562

Demandé le 3 octobre 1931 — Délivré le 3 novembre 1932

P. 1, lines 1-2
"La présente invention a pour objet un procédé d'obtention de l'oxyde d'éthylène . . ."

P. 1, lines 8-11
". . . des réactions de base du procédé qui peuvent être exprimées sous la forme suivante:

P. 1, lines 1-6
"La présente invention a pour objet un procédé d'obtention de l'oxyde d'éthylène qui consiste principalment à soumetre l'éthylène à l'action simultanée de l'oxygène de l'air et de l'eau en présence d'un catalyseur et d'hydrogène."

P. 1, lines 12, 16-17
"Cette réaction . . . à une température comprise entre 150 et 400° C."

P. 1, lines 18-19, 23
"Les catalyseurs employés peuvent être les suivants:
"2° L'argent seul;"

P. 1, lines 46-49
"L'éthylène peut provenir de n'importe quelle source: gaz des fours à coke, hydrogénation de l'acétylène, déshydratation de l'alcool éthylique, et autres."

P. 2, lines 4-5
"L'oxygène peut être l'oxygène de l'air, ce dernier gaz étant introduit dans la réaction."

P. 2, lines 32-37
"Dans un tube, par exemple en silice, on introduit de l'argent en poudre, ou sous forme de toile, sur un support ou non et, après avoir porté le tube à une température comprise entre 150 et 400°. . . ."

1st Addition—No. 41,811

Demandée le 19 décembre 1931 — Délivrée le 21 janvier 1933

P. 1, lines 5-8
". . . consiste principalement à soummettre l'éthylène à l'action simultanée de l'oxygène et de l'eau en présence d'un catalyseur . . ."

P. 1, lines 11-16
". . . la réaction a lieu . . . en présence de catalyseurs parmi lesquels est cité l'argent sous forme colloïdale, sous forme de poudre ou de toile."

P. 1, lines 28-33
"c. On augmente le rendement de la réaction en diminuant le CO^2 qui se forme, par l'introduction, dans cette réaction, d'une quantité d'eau convenable et en mélangeant au préalable, aux gaz réagissants, un volume convenable de CO^2;"

P. 2, lines 2-3
"Les aldéhydes formées ne le sont qu'à l'état de traces."

P. 2, lines 11-16
"De plus, on a constaté que la réaction donnant le CO^2 est, contrairement à ce que l'on a cru jusqu'à ce jour, une réaction d'oxydation indépendante de celle qui donne l'oxyde d'éthylène et de celle qui donne les aldéhydes."

P. 2, line 24

"$C_2H_4 + 3O_2 = 2\ H_2O + 2\ CO_2$"

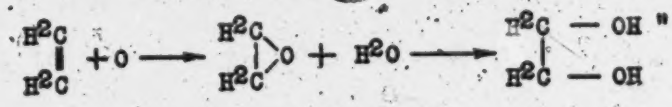
French Patent No. 729,952

Demandé le 27 mars 1931 — Délivré le 3 mai 1932

P. 1, lines 3-11

". . . à soumettre l'éthylène provenant de n'importe quelle source: gaz des fours à coke, hydrogénation de l'acétylène, déshydratation de l'alcool éthylique, etc., à l'action simultanée de l'oxygène de l'air et de l'eau.

"La réaction étant à la base de ce procédé peut être envisagée de la façon suivante:



"

P. 1, lines 45-47

"1° Procédé d'obtention de l'éthylèneglycol par action simultanée de l'oxygène de l'air et de l'eau sur l'éthylène; . . ."

2nd Addition—No. 41,474

Demandée le 10 septembre 1931 — Délivrée le 25 octobre 1932

P. 1, lines 11-22

"La présente addition a pour but de préciser que l'obtention du glycol qu'on obtient à partir de l'oxydation de l'éthylène par l'oxygène de l'air, en présence d'eau, provient nécessairement de la saponification de l'oxyde d'éthylène obtenu en première phase par oxydation directe de l'éthylène. Le mécanisme, indiqué dans le brevet principal et la première addition relative à celui-ci, tend donc à produire au préalable l'oxyde d'éthylène qui, par saponification directe, donne le glycol."

French Patent No. 729,952

P. 1, lines 6-7

". . . à l'action simultanée de l'oxygène de l'air et de l'eau."

P. 1, lines 31-34

"Ces deux gaz seront dirigés dans un tube de réaction . . . contenant un des catalyseurs indiqués ci-dessus, . . ."

P. 1, lines 52-53

"3° La température de réaction est maintenue de préférence entre 150 et 300° C.;"

2nd Addition—No. 41,474

P. 1, lines 33-38

"Dans un tube, par exemple en silice, on introduit de l'argent en poudre, ou sous forme de toile, sur un support ou non et, après avoir porté le tube à une température comprise entre 250 et 400°, . . ."

P. 2, lines 17-23

"2° Obtention d'oxyde d'éthylène par oxydation directe de l'éthylène par l'oxygène de l'air en présence de vapeur d'eau et en contact des catalyseurs cités dans le brevet et la première addition, notamment l'argent et ses mélanges avec le bismuth, l'antimoine, le plomb, l'étain, le nickel."

French Patent No. 729,952

P. 2, lines 5-7

"5° En évitant d'ajouter de l'eau à la réaction on peut obtenir uniquement de l'oxyde d'éthylène."

4th Addition—No. 41,810

Demandée le 19 décembre 1931 — Délivrée le 21 janvier 1933

P. 1, lines 27-32

"c. On augmente le rendement de la réaction en diminuant le CO_2 qui se forme, par l'introduction, dans cette réaction, d'une quantité d'eau convenable et en mélangeant au préalable, aux gaz réagissants, un volume convenable de CO_2;"

P. 1, lines 45-46

"Les aldéhydes formées ne le sont qu'à l'état de traces."

P. 2, lines 8-13

"De plus, on a constaté que la réaction donnant le CO_2 est, contrairement à ce que l'on a cru jusqu'à ce jour, une réaction d'oxydation indépendante de celle qui donne l'oxyde d'éthylène et de celle qui donne les aldéhydes."

P. 2, line 21

"$C_2H_4 + 3O_2 = 2\ H_2O + 2\ CO_2$"

Plaintiff's Exhibit 21.

DISTRICT COURT OF THE UNITED STATES

FOR THE DISTRICT OF MARYLAND

CARBIDE AND CARBON CHEMICALS CORPORATION, Plaintiff, vs. U. S. INDUSTRIAL CHEMICALS, INC., Defendant.	Civil Action File No. 249

Plaintiff's Statement Regarding its Operations Under Lefort Reissue Patent No. 20,370

In its manufacturing plant at South Charleston, West Virginia, plaintiff is now and has continuously since November, 1937, engaged in the commercial production of ethylene oxide by direct catalytic oxidation of ethylene with molecular oxygen. In doing so, plaintiff employs a chamber containing a catalyst essentially composed of silver disposed on an inert carrier. A gaseous mixture containing ethylene and the constituents of atmospheric air is passed continuously and substantially uniformly for extended periods of time through this chamber in intimate contact with said catalyst.

The gaseous mixture enters the chamber under superatmospheric pressure and it is saturated with water vapor at the working pressure and at approximately atmospheric temperature.

Plaintiff's Exhibit 21 (continued).

Throughout the operation the temperature within the said chamber is maintained substantially constant at a temperature between 150° and 400° C.

The ethylene and oxygen in the gaseous mixture react within the chamber, in the presence of the catalyst and of water, to produce ethylene oxide in substantial amount; and the resultant gaseous mixture, containing the said oxide and no substantial amounts of aldehydes, passes out of the said chamber continuously during the operation. The said resultant gaseous mixture contains usually about 1.5 per cent to about 2.0 per cent by volume of water vapor in addition to that contained in the entering gas mixture.

Thereafter, the ethylene oxide is dissolved out of the said resultant gaseous mixture and recovered in liquid form.

L. A. WATSON
Counsel for Plaintiff

Dated: May 21, 1940.

Plaintiff's Exhibit 22.

Munson Deposition Exhibit 8 for Identification
May 3, 1940
Stanley J. Cohara, Notary Public

THE DISTILLERS CO. LTD.

TELEPHONE WHITEHALL 1040.
(9 LINES)

TELEGRAMS -
"FERMENTER, LESQUARE, LONDON"

KINNAIRD HOUSE,
2, PALL MALL EAST,
LONDON, S.W.1. December 6th 1935.

C.E. Adams Esq.,
Air Reduction Company
Lincoln Building,
42nd Street,
NEW YORK.

Dear Adams

Many thanks for your letter of November 26th enclosing your Report on Research on Alcohol etc. for the period July 1 to October 1 of this year.

I send you herewith our Research Report for the period April 1st to December 1st 1935, direct as arranged and shall be very glad to answer any question you may have to ask in connexion with it.

With kind regards,
Yours sincerely,

Thos Ansard

Plaintiff's Exhibit 22 (continued).

3.

Work on the actual hydration of ethylene and propylene has been brought to a close on the small plant which has been in constant operation for two years. Sufficient information has been obtained regarding the activity of the solid catalysts employed, their active life and their mechanical stability. When this plant was dismantled recently, no corrosion of any part of the plant could be found.

Ethylene oxide.

Some time ago this company became interested in a French catalytic process for the manufacture of ethylene oxide from ethylene. The work at the time had not proceeded far but the process and methods leading up to industrial production have since been developed in these laboratories. Using air with 5% ethylene and working at ordinary atmospheric pressure and at temperatures of 300° to 350°C, a conversion of ethylene as high as 50% has been obtained, with an efficiency figure of 50 and a conversion figure of 50, (by which is meant that half the ethylene present is utilised per passage, half going to carbon dioxide and half going to ethylene oxide.) At present a unit comprising 10 catalytic tubes is being operated experimentally, and designs are being prepared for a larger unit which is expected to yield about 1 ton of ethylene per week. Work is in progress on the separation of the ethylene oxide from the resultant gaseous mixture of carbon dioxide and ethylene oxide and means of condensing olefine oxides and organic hydroxy compounds are being worked out, using as catalysts, for example, tertiary aliphatic amines. The conditions employed are for example 60-100°C and say 5-10 atmospheres pressure, under which conditions the condensation seems to proceed satisfactorily.

Lacquers.

The preparation of brushing lacquers is being studied, using nitrocellulose, water and solvents (a) where the water is the continuous phase, and (b) where the solvent is the continuous phase. With the former kind the results have not proved satisfactory, as the film, although continuous,

Plaintiff's Exhibit 22 (continued).

RADIOGRAM

Mackay Radio

ADDRESS: 67 Broad Street, New York

December 11, 1935

Board
Fermenter
Lesquare
London

PARTICULARLY ANXIOUS TO GET YOUR FULL DATA AT EARLIEST POSSIBLE DATE RE ETHYLENE OXIDE ABOUT WHICH EYRE SPOKE TO METZGER.

ADAMS

AIR REDUCTION SALES CO.

TO SEND A MESSAGE TELEPHONE WHITEHALL 4-6789 OR ANY POSTAL TELEGRAPH OFFICE. FOR OTHER PURPOSES — TELEPHONE BOWLING GREEN 9-3890. FOR MESSENGER TELEPHONE POSTAL TELEGRAPH OR RING POSTAL CALL BOX.

Plaintiff's Exhibit 22 (continued).

RCA

RADIOGRAM

RCA COMMUNICATIONS INC

RECEIVED AT ... NEW YORK, AT ... 9 26 ... STANDARD T

LX34 XUX GB2717

LONDON 43 13 1221

ADAMS DENATURED

NEWYORK

YOUR CABLE RECEIVED STOP FURTHER INFORMATION CONTAINED IN RESEARCH REPORT POSTED DECEMBER SIXTH STOP WE HAVE BRITISH EMPIRE RIGHTS ONLY STOP YOU SHOULD NEGOTIATE DIRECT WITH SOCIETE FRANCAISE DE CATALYSE GENERALISE 26 RUE PIERRE L HONNE COURBEVOIE SEINE FRANCE

BOARD

TELEPHONE HAnover 2-1811 To secure prompt action on inquiries, this original RADIOGRAM should be presented at the office R. C. A. COMMUNICATIONS, Inc. In telephone inquiries quote the number preceding the place of o

Plaintiff's Exhibit 22 (continued).

December 17, 1935

Mr Thomas H Board
The Distillers Co Ltd
Kinnaird House
2, Pall Mall East
London, S W 1
England

Dear Board

Thank you so much for your wire of December 13, and for your letter of December 6, enclosing your report on Research on Alcohol, for the period July 1 to October 1 of this year.

I have gone over this report with Metzger, and he has written me a memorandum, copy of which is enclosed. Any further information you can give us, as indicated by this memorandum, would be greatly appreciated.

With best regards,

Sincerely yours,

President

CEA:MMC

Enclosure

22/5

Plaintiff's Exhibit 22 (continued).

INTERIOR LETTERHEAD

DELIVER THIS LETTER TO	ADDRESS REPLY TO
Mr. C E Adams	Mr. F J Metzger
At	At

Date Dec 17,1935

SUBJECT:- PROGRESS REPORT FROM THE RESEARCH AND DEVELOPMENT DEPARTMENT OF THE DISTILLERS COMPANY,LIMITED April 1, 1935 to December 1, 1935

Replying to your letter dated

We have all been very much interested in this report.

Oil Cracking

Their experiences with oil cracking are substantially in line with our own. However, we have an advantage in our experimental work in that we can secure definite types of oils and thereby control our products to better advantage.

Cellulose Triacetate

We shall be glad to receive from time to time further information on their cellulose triacetate by the Berl process, particularly any developments in finding new solvents and plasticizers for this product.

Ethylene Oxide

We are of course interested in their work on ethylene oxide. The report is a little bit ambiguous and subject to more than one interpretation. For instance, it is not quite clear whether 1 1/4 or 2 1/2% of the 5% ethylene entering the reaction chamber is converted into ethylene oxide.

We also would be very glad to get more information on this subject, such as details of furnace construction, method of preparing the catalyst and mode of operation of the unit, whenever such information is available.

F J Metzger

FJM:IH

Plaintiff's Exhibit 22 (continued).

COPY.

THE DISTILLERS COMPANY LIMITED.

Research Department.

14th January 1936.

T.H. Board Esq.,
The Distillers Co. Ltd.,
etc.

Dear Mr. Board, Ethylene Oxide.

I see from Dr. Metzger's memorandum dated 17th December that he has not found very clear the figures which I gave in my report dated 6.12.35. under the heading of "Ethylene Oxide".

It is a little difficult to put briefly into words, but in amplification I may say that my meaning was that when using a 5% concentration of ethylene, half of the ethylene per passage remains unchanged, the other half being converted to an equal extent into ethylene oxide and carbon dioxide. This means that one quarter of the 5% original ethylene is used up in the formation of ethylene oxide and a quarter appears as carbon dioxide.

Since then, however, by modifying the apparatus, improved yields have been obtained and our present results may be stated as follows: Using an air mixture containing 5% ethylene, we are getting 75% of the ethylene utilised per passage, 45-50% of such ethylene utilised going to the formation of ethylene oxide, which means that 35-37% of the ethylene used appears as ethylene oxide per passage. This does not, however, represent the best results we have obtained, but they are figures upon which we fell we can rely. Our best figures have been 85% utilisation of ethylene and 55% of ethylene utilised going to produce ethylene oxide. I may say that the reaction gives a remarkably clean product. Only by chemical means can one detect by-products such as acetaldehyde, acids and per-compounds, presumably per-acetic acid. These improved results have justified modifying the design of the small plant which was being arranged for, but I hope that before long we shall be in a position to produce ethylene oxide regularly on a small scale.

With .....

Plaintiff's Exhibit 22 (continued).

With regard to details of plant and catalyst, I do not think we are free to communicate these to other parties for the following reasons: (1) We are working the problem out in conjunction with the Société Française de Catalyse Généralisée; (2) we are only interested in British Empire rights, and (3) although we are under an obligation to exchange with the S.F.C.G. all details of catalyst preparation and plant construction, the French firm will have to negotiate foreign rights.

I am returning the letter and memorandum as promised.

Yours truly,

(sd) J. Vargas Eyre.

Plaintiff's Exhibit 22 (continued).

HE DISTILLERS CO LTD.

TELEPHONE-WHITEHALL 1040
(5 LINES)

TELEGRAMS -
"FERMENTER, LEDQUARE, LONDON"

KINNAIRD HOUSE,
2, PALL MALL EAST,
LONDON, S.W.1. January 17th, 1936.

C.E. Adams Esq.,
Air Reduction Company,
Lincoln Building,
42nd Street,
NEW YORK CITY. U.S.A.

Dear Adams,

I am sorry to have been so long in sending you an answer to your letter of December 17th, but I have been very busy.

I have discussed the Progress Report with Dr. Eyre particularly regarding Ethylene Oxide, and I enclose herewith copy of his reply.

With reference to Cellulose Triacetate, I would like to draw your attention to the fact that we put you in direct touch with Dr. Berl for his Cellulose Triacetate Process. We have the British rights only, but you can get full information from Dr. Berl on the Process and the Solvents and Plasticisers required. At the moment our small commercial experimental plant is in course of erection and is not likely to be working for a couple of months yet.

Yours sincerely,

Thos Board

P.S. Berl's address is the Carnegie Institute Pittsburg University.

My people are very interested in Cyanacetic Anhydride acid - have you any information on the subject?

22/a

Plaintiff's Exhibit 22 (continued).

January 29th
1936

Thomas H. Board, Esq.
The Distillers Co. Ltd
Kinnaird House
2, Pall Mall East
London, S.W. 1
England

Dear Mr. Board:

Due to Carl Adams being in Florida, your letter of January 17th regarding Ethylene Oxide has been turned over to me.

Your experiments look very interesting. In case we should deem it advisable to get in touch with the Societe Francaise de Catalyse Generalisee about this situation, I would like your advice as to how this had better be done; would it be best to start this through you, approach them direct, or have they representatives in the U.S.A.? We would appreciate any help you can give us on this and will write you later in reference to it.

Regarding synthetic proprionic acid, we have no direct information about this in our own Research end, but I believe DuPont is making this from normal Propyl which they obtain as a by-product in their Methyl Alcohol Synthesis.

With all best wishes, I am

Faithfully yours,

CSM/ed

Plaintiff's Exhibit 22 (continued).

THE DISTILLERS CO. LTD.

TELEPHONE: WHITEHALL 1040 (6 LINES)

TELEGRAMS: "FERMENTER, LESQUARE, LONDON"

KINNAIRD HOUSE,
2, PALL MALL EAST,
LONDON, S.W.1.

10th February 1936

C.S. Munson, Esq.,
U.S. Industrial Alcohol Co.,
60, East 42nd Street,
NEW YORK.

Dear Munson,

Ethylene Oxide

In reply to your letter of the 29th January re ethylene oxide, the Societe Francaise de Catalyse Generalisee have no agent in the States and so I would write to them direct (at 26, Rue Pierre l'homme, Courbevoie, Seine) and not through us. I know they are in treaty with other people in America for the sale of their rights, but I do not believe anything has been completed.

I am much obliged for what you say about synthetic propionic acid, which I will take up with Dr. Eyre.

With kind regard,

Yours sincerely,

Howard

22/11

Plaintiff's Exhibit 22 (continued).

February 26th, 1936

Mr. Eugene W. Stetson
The Guaranty Trust Co.
140 Broadway
New York City

Dear Gene:

I am enclosing a letter addressed to the Societe Francaise de Catalyse Generalisee, who have a process for making Ethylene Oxide, which is turned into Ethylene Glycol (the basis of Prestone). From our various discussions, you know why we are interested in this process.

This information has come to us through the Distillers Co. Ltd. of England, who suggest we approach the French Company direct; as we are approaching them "cold" we felt it would be better to have it go through the Guaranty Trust, if such a procedure is agreeable to you. We are doing it in this way as we understand the French Company are negotiating with other people in the United States covering this process. If you would not mind asking your Paris Branch to handle this, it would be much appreciated by us.

Faithfully yours,

CSM/cd

Plaintiff's Exhibit 22 (continued).

Munson Deposition Exhibit 1 for Identification
May 3, 1940
Stanley J. Cohara, Notary Public.

February 26th
1936

Societe Francaise de Catalyse Generalisee
26, Rue Pierre L'homme
Courbevoie
Seine

Dear Sirs:

Our good friends the Distillers Co. Ltd. of England have suggested to us that you have a process for the making of Ethylene Oxide which might be of interest to this Company. We would be interested in such a process and would be glad to discuss this with you.

I am sending this letter to you through our good friends The Guaranty Trust Company, who will give you information about our Company and to whom we would be glad to have you give any information that you care to in reference to this process and in reference to any interest you might have in developing this in the United States.

Faithfully yours,

C. S. MUNSON
President

Plaintiff's Exhibit 22 (continued).

Guaranty Trust Company of New York

A merger of
National Bank of Commerce in New York - Guaranty Trust Company of New York
Organized 1839 Organized 1864

140 Broadway

Capital $90,000,000 Surplus $170,000,000.

Member of Federal Reserve System

New York Offices
140 Broadway
Cable Address: Guarantrust
Fifth Avenue at 44th St
Cable Address: Satoshin
Madison Avenue at 60th St
Cable Address: Guarantrust

Foreign Department
Kent G. Colwell
Assistant Manager

Foreign Offices
Cable Address: Guaran

London
Liverpool
Brussels

Paris
Havre
Antwerp

New York February 26, 1936.

In replying please refer to
504-20

Mr. C. S. Munson, President,
U. S. Industrial Alcohol Co.,
60 East 42nd Street,
New York, N.Y.

Dear Mr. Munson:-

Mr. Stetson, who is leaving town tomorrow, has asked me to forward the letter addressed to the Societe Francaise de Catalyse Generalisee, which was enclosed in your letter to him of today's date.

Your enclosure has been sent to our Paris Office with a covering letter mentioning our close and important relations with your good company, which we request them to mention when delivering your letter to the Societe Francaise de Catalyse Generalisee.

It is a pleasure for us to be of service in this way and, if there is anything further in this connection that you would like to have us do, it will be a privilege for me to follow the matter on your behalf.

Yours very truly,

K. G. Colwell

Assistant Manager.

Plaintiff's Exhibit 22 (continued).

February 28th
1936

Mr. Kent G. Colwell,
Assistant Manager,
Foreign Department
Guaranty Trust Co. of New York
140 Broadway
New York City

Dear Mr. Colwell:

Many thanks for your very kind letter of February 26th and for your kindness in forwarding the letter to the Societe Francaise de Catalyse Generalisee through your Paris office.

Faithfully yours,

C. S. Munson
President.

CSM/cd

Cross Reference in Guaranty Trust Co file
See also Miscellaneous "C" file for correspondence with Societe Francaise de Catalyse Generalisee

2216

Plaintiff's Exhibit 22 (continued).

Guaranty Trust Company of New York

A merger of
National Bank of Commerce in New York - Guaranty Trust Company of New York
Organized 1929 Organized 1864

140 Broadway

Capital $90,000,000. Surplus $170,000,000.

Member of Federal Reserve System

"New York Offices
140 Broadway
Cable Address: Palmbeach
5th Avenue at 44th St.
Cable Address: Palmave
Lexington Avenue at 60th St.
Cable Address: Lexmade"

Foreign Department
Kent G. Colwell
Assistant Manager

Foreign Offices
Cable Address Garuaran

London
Liverpool
Brussels

Paris
Havre
Antwerp

New York March 23, 1936.

In replying please refer to
504-20

Mr. C. S. Munson, President,
U. S. Industrial Alcohol Co.,
60 East 42nd Street,
New York, N.Y.

Dear Mr. Munson:-

With reference to our telephone conversation this afternoon, a copy of letter from our Paris Office and of the memorandum enclosed with it are herewith enclosed, in connection with your letter to the Ste. Francaise de Catalyse Generalisee delivered by our Paris Office.

Let us urge you once more to make use of us in any way possible if you feel that we can render any additional service in this matter.

Very sincerely yours,

K. G. Colwell
Assistant Manager.

Enclosure.

Plaintiff's Exhibit 22 (continued).

GUARANTY TRUST COMPANY OF NEW YORK

Paris Office

Letter to HEAL office Foreign Department,

Attention of: Mr. Kent G. Colwell, Assistant Manager.

PARIS 11th, March 1936.

U. S. Industrial Alcohol Co.
Ste. Franç. de Catalyse Generalisee

Dear Kent,

Upon receipt of your letter of Feb. 27th, enclosing a copy of the letter from President Munson of the United States Industrial Alcohol Co. to the Ste. Francaise de Catalyse Generalisee, we had the delivery made by Mr. d'Algay, with whose capacity and personality you are doubtless familiar. From the attached report you will see that the Managing Director of the Societe Francaise de Catalyse Generalisee is replying direct to the United States Industrial Alcohol Co. Needless to say, Mr. d'Algay went fully posted as to the standing of the United States Industrial Alcohol Co. and our relationship with them.

With kind regards,

Yours sincerely,

(Signed) ELLIOTT H. LEE
Vice President

EHL/CPG Encl.

Munson Deposition Exhibit 2 for Identification
May 3, 1940
Stanley J. Cohara, Notary Public

22/17

Plaintiff's Exhibit 22 (continued).

March 11, 1936.

U.S. INDUSTRIAL ALCOHOL COMPANY

Call on Mr. Francon, Managing Director of
SOCIETE FRANCAISE LE CATALYSE GENERALISEE

Mr. F. knows the U.S. Industrial Alcohol Company and Distillers Company Ltd., of England, the latter being the licencee of his process for the manufacture of ethylene oxide.

Mr. F. is very much interested in selling the licence for his process particularly in the United States which, in his opinion, represent several times the possibilities of production of all the rest of the world. He points out, however, that this transaction requires careful consideration on his part because he wants to make the most of this licence and will sell it only to the Companies which are best qualified to commercially give it the greatest development in the United States. This involves technicalities which can only be discussed with persons conversant with chemistry. He suggests, therefore, that the subject Company should send somebody to see him on the matter thinking that said Company may have some technical representative in Europe.

He has been investigating the matter in respect to possibilities in the United States for over one year and has been approached by four important Companies connected with producers of ethylene gas (cokeries petroleum refining). He considers himself now as well posted up in respect to United States possibilities and now anxious to bring the matter to a conclusion within the three or four next months.

In Europe, he has sold the licence in several important countries (England, Belgium, Italy) and quite recently to the U.R.S.S. He has also been approached by Germany which wanted to buy the licence for all the countries in which it has not yet been sold, but this overture has been declined.

In short, Mr. F. who knows english quite well, is going to reply direct to the U.S. Industrial Alcohol Company and suggests that we write also on our side but is positive that nothing useful

22/10

Plaintiff's Exhibit 22 (continued).

will be achieved unless some direct contact is established between a representative of the subject Company and himself to discuss technicalities and terms.

* * *

Ethylene is a by-product of cokeries and petroleum cracking; it can be manufactured from alcohol but the cost price if probably higher. Ethylene oxide has direct uses and is also an important intermediary in the manufacture of solvents, war gas, etc. 22/1.9

Plaintiff's Exhibit 22 (continued).

SOCIÉTÉ FRANÇAISE DE CATALYSE GÉNÉRALISÉE

Follow up 3/9/36

PROCÉDÉS DE MONSIEUR LE PROFESSEUR CHARLES HENRY

Docteur honoraire de l'École des Hautes Études

Anonyme au Capital de 6 000 000 de Francs

SIEGE SOCIAL TRANSFERE
AU LABORATOIRE
20, Rue Florent Lhomme
COURBEVOIE (Seine)
Tél. : DEFense 06-56

8, Avenue Percier
PARIS (VIIIe)

Courbevoie le 13 Mars 1936.

U.S. Industrial Alcohol Cº

60 East 42nd Street

New York

Messieurs ,

Nous avons l'honneur de vous accuser réception de votre lettre du 26 Février ,que nous a remise vos amis The Guaranty Trust CY.

Nous ne demandons pas mieux que d'envisager une possibilité d'affaire avec votre Société pour les U.S. ,mais nous estimons qu'il est absolument indispensable avant toute chose que notre Société puisse avoir un contact technique avec un de vos représentants qualifié à cet effet .

Pensant que votre opinion sera conforme à la notre en cette circonstance ,nous vous prions ,Messieurs , d'agréer , nos salutations distinguées .

L'Administrateur
Directeur Général

Munson Deposition Exhibit 3 for Identification
May 3, 1940
Stanley J. Cohara, Notary Public

Plaintiff's Exhibit 22 (continued).

March 24th
1926

Mr. Kent G. Colwell, Assistant Manager
Foreign Department
Guaranty Trust Company of New York
140 Broadway
New York City

Dear Mr. Colwell:

Many thanks for your very kind letter of March 23rd and for all the trouble you and your Paris office have taken in connection with our communications with the Ste.Francaise de Catalyse Generalisee. This is very much appreciated by all of us.

Faithfully yours,

C.S.Munson

CSM/cd

22/21

Plaintiff's Exhibit 22 (continued).

THE DISTILLERS CO. LTD.

TELEPHONE WHITEHALL 1040 (6 LINES)

TELEGRAMS "FERMENTER, LESQUARE, LONDON"

KINNAIRD HOUSE,
2, PALL MALL EAST,
LONDON, S.W.1.

April 16th 19[illegible]6.

C.E. Adams Esq'.,
Air Reduction Company,
Lincoln Building,
42nd Street
NEW YORK CITY. U.S.A.

Dear Adams,

I enclose herewith our Progress Report from the Research Department for the period ending 1st March, together with copy of a letter which Dr. Eyre has written to me.

We shall be very glad to answer any questions you may like to ask on the Report.

With kind regards,
Yours very sincerely,

Thos Board

Plaintiff's Exhibit 22 (continued).

3.

Hydration of olefines: No further experimental work has been done in so far as ethylene and propylene are concerned, attention having been directed to the hydration of butylene and higher olefines in the liquid phase at relatively low temperatures and pressures. As a result a small plant for continuous operation is being designed to test the possibilities of liquid phase hydration as applied to butylene, butadiene and amylenes, substances which are known to be formed as a result of cracking oil at the moderate temperatures of 700-750°C.

Ethylene oxide: The work on ethylene oxide has been continued with fair success and a larger plant estimated to produce about 1½ to 2½ kg. of ethylene oxide per day has been erected and is shortly coming into use. A good deal of difficulty has been experienced in getting suitable material from which to make the active catalyst, but it is expected to have this plant under way shortly. Satisfactory means have been worked out using charcoal for adsorbing and desorbing the ethylene oxide produced. The recovery of ethylene oxide, both in the form of ethylene oxide and glycol, seems to vary between 92% and over 100% of the ethylene oxide introduced. The amount of ethylene glycol recovered as such varies between 0.5 and 1.8% of the ethylene oxide used. This represents a 96.5% recovery of ethylene oxide and a total recovery as oxide + glycol of 97.4%.

Glycol ethers: Considerable work has been done on the direct conversion of ethylene oxide to glycol ethers, not only by the employment of tertiary bases as catalysts but when using certain solid catalysts. The best method so far arrived at consists in passing continuously a mixture of ethylene oxide with the appropriate alcohol over a chosen solid catalyst at temperatures of about 100°C and pressures of 5-10 atmospheres. In this way one is able continuously to prepare the desired glycol ether in a state of great purity, and containing only about 15% of the less desirable ethers of poly glycols. Present results show about 95% utilisation of ethylene oxide per passage over the catalyst.

LACQUERS:

Very little further work has been done on this subject, pending information as to the possible industrial application of lacquers to metal, wood and plaster board.

CELLULOSE TRIACETATE:

The demonstration plant referred to in the last report is now nearing completion, and the only experimental work being done in this particular field has reference to the application of cellulose triacetate for industrial purposes, which has necessitated a careful study of the question of solvents and plasticisers. In this connection it has been found, by very simple means, possible to prepare bottle caps having cellulose triacetate as a base instead of nitro-cellulose. In so far as these are able to carry pigments and

22/33

Plaintiff's Exhibit 22 (continued).

Munson Deposition Exhibit 4 for Identification
May 3, 1940
Stanley J. Cohara, Notary Public

April 29, 1936

Societe Francaise De Catalyse Generalisee
26 Rue Pierre Lhomme
Courbevoie (Seine)
France

Gentlemen:

We wish to acknowledge and thank you for your letter of March 12, 1936, and for your expression of good will toward our Company.

We are very much interested in a satisfactory process for the production of ethylene glycol and have looked into your U. S. Patent No. 1,998,078, as well as some of your French patents on the same subject. A study of these patents leaves some doubt in our mind as to whether your process as disclosed could compete with the well-known chlorhydrin process operated on a large scale in the United States at the present time.

It would help us very much to come to an early conclusion with respect to the applicability of your process in the United States and the advisability of sending one of our engineers to Paris, if you could tell us whether or not you have materially improved the process as outlined in the above mentioned patents; also whether either you or your licensees have carried out the operation on a sufficiently large scale to enable you to establish accurate cost data. We would like very much to know the largest scale on which your process has been operated to date and at what price you believe ethylene glycol could be manufactured, starting with pure ethylene at a cost of 2 1/2 cents per pound.

We would greatly appreciate an early reply, and beg to remain

Cordially yours,

LB

C. S. Munson
President

Plaintiff's Exhibit 22 (continued).

Museum Deposition Exhibit 5 for Identification
May 2, 1940 — Stanley J. Cohara, Notary Public

SOCIÉTÉ FRANÇAISE DE CATALYSE GÉNÉRALISÉE

PROCÉDÉS DE PROFESSEUR CHARLES HENRY

SIEGE SOCIAL TRANSFERE
AU LABORATOIRE
26, Rue Pierre Lhomme
COURBEVOIE (Seine)
Tél. : DÉFense 0049

11, Avenue Percier
PARIS (VIIIe)

Courbevoie le 11 Mai 1936

Industrial Alcohol C°

50 East, 42ND Street

New-York

Messieurs,

Nous avons l'honneur de vous accuser réception de votre lettre du 29 Avril, dont nous vous remercions.

Malheureusement pour nos rapports futurs, nous sommes dans l'obligation de vous dire que [redacted] les tractations que nous avions aux Etats-Unis, avant d'être en contact avec votre Compagnie, ont recemment abouti, et que nous avons cédés tous nos droits exclusifs pour les états-unis, à la Carbide and Carbon Chemical Corporation.

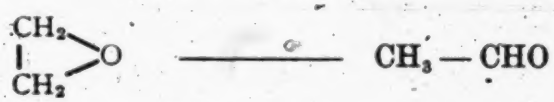
Nous espérons bien vivement que nous aurons l'occasion dans d'autres domaines de faire des accords futurs avec votre Compagnie, et avec nos regrets nous vous envoyons, Messieurs, l'expression de nos sentiments les meilleurs.

L'Administrateur
Directeur Général

Plaintiff's Exhibit 25.

BERICHTE (1903) p. 2017

Ethylene oxide, under the influence of aluminum oxide, was isomerized to acetaldehyde.



Plaintiff's Exhibit 27.

Translation of German Patent No. 547,641

Rheinische Kampfer-Fabrik G. M. b. H. in Düsseldorf-Oberkassel*)

*) The patentee as given by the patent solicitor is Dr. Josef Kathol in Düsseldorf-Oberkassel.

"Process for the Preparation of Acetaldehyde"

Patented in Germany as of December 10, 1930
Published March 29, 1932

* * * * *

On the other hand the surprising fact was established that silica gel is an outstanding catalyst for the transformation of ethylene oxide into acetaldehyde.

* * * * *

Plaintiff's Exhibit 28 for Identification.

FORMATION OF WATER

SUMMARY:

An experiment was carried out to investigate the rate of production of ethylene oxide and of carbon dioxide and water when ethylene and dried air were passed over a silver catalyst at a temperature within the range of 150-400° C. So far as could be determined formation of all of these compounds started simultaneously as soon as the reactants met the catalyst. Ethylene oxide and carbon dioxide plus water were produced throughout practically the entire length of the catalyst section.

EXPERIMENTAL:

These experiments were carried out in a 1-inch, oil-jacketed steel tube heated for a length of 36 inches. Temperatures were maintained at about 210° and recorded through a steel thermocouple well leading into the oil employed as the heat transfer medium. A stainless steel tube ⅛" in outside diameter extended through the center of the catalyst bed. The stainless steel tube was movable and was drawn down through the catalyst bed so that spot samples of the reaction products could be collected at various points in the catalyst mass.

The catalyst was prepared by evaporating to dryness an aqueous mixture of 97 grams of silver oxide and 250 cc. of 4 x 8 mesh carborundum. Two hundred cc. of this product were placed in the lower section of the tube mak-

Plaintiff's Exhibit 28 for Identification (continued).

ing a section 14½" long. The upper part of the tube was packed with inert material and served as a preheater. An air-ethylene mixture containing 5% ethylene was fed at the rate of 50 liters per hour. The air used was first passed through soda lime and calcium chloride to remove carbon dioxide and water.

In each instance, gas samples were collected from the exit end of the tube (Sample A) and from the stainless steel tube at various points in the catalyst bed (Sample B). The yields and efficiencies reported in the appended table were calculated from the results of analyses of these samples. Samples A were taken to show that the differences in composition of Samples B were due to the location in the catalyst bed and not to variations of the conditions during the run.

Qualitative tests for carbon dioxide were made by passing the gas through barium hydroxide solution and for ethylene oxide by the method of Deckert (Z. Angew. Chem. *45*, 559-62). Negative tests for ethylene oxide and carbon dioxide were obtained from the gas mixture just above the catalyst. Positive tests for both compounds were obtained just below the top of the catalyst indicating that these compounds were formed simultaneously. Additional quantitative data are given in the table.

The results prove that the production of ethylene oxide and of carbon dioxide and water continues throughout the length of the catalyst section, and that the ethylene oxide, carbon dioxide and water formed each gradually increases as the gas mixture is passed over an increasing length of catalyst bed.

Plaintiff's Exhibit 28 for Identification (continued).

Sample A		Sample B			
Single Pass Yield %	Eff. %	Single Pass Yield %	Eff. %	Ratio: Ethylene Oxide / Water	Sample Collected
19	57	1.4	44	0.41	½" below top of catalyst
21	62	2.5	58	0.70	1" below top of catalyst
21.5	65	3.4	56	0.63	1½" below top of catalyst
21	63	4.3	60	0.80	2" below top of catalyst
22	66	6.2	63	0.87	3" below top of catalyst
20.8	66	7.6	62	0.83	4" below top of catalyst
20.4	65	8.8	62	0.83	5" below top of catalyst
20.4	65	12.2	66	1.0	7" below top of catalyst
21.0	64	17.1	68	1.11	10" below top of catalyst
21.0	65	19.7	67	1.04	13" below top of catalyst
21.7	67	21.7	67	1.04	Just below bottom of catalyst

5-23-40

SUPPORTS

SUMMARY:

A series of one-day tests was carried out in which the following substances were used as supports for silver catalysts employed in the oxidation of ethylene to ethylene oxide: glass, fire clay, porcelain, asbestos, infusorial earth, Zeolite, magnesium oxide, carborundum, copper, zinc, pumice, Alundum, Filtros, barium sulfate and brick. Ethylene oxide was obtained with each.

A table of data is appended giving typical results obtained with each support.

In addition to these one-day tests, catalysts supported on carborundum, Alundum and Filtros were tested for several days. In each case the efficiency rose to above 60%.

EXPERIMENTAL:

These experiments were carried out in a 1-inch oil-jacketed, steel tube heated for a length of 36 inches.

Plaintiff's Exhibit 28 for Identification (continued).

Temperatures were recorded through a ¼-inch steel thermocouple well extending through the center of the tube. The catalyst was placed in the lower section of the tube surrounding the thermocouple well. The upper part of the tube was packed with inert material and served as a preheater. An ethylene-air mixture, usually containing about 5% ethylene, was fed at the rate of 50 liters per hour.

The catalysts were prepared by evaporating to dryness an aqueous mixture of 200 cc. of support and 77 grams of silver oxide. When possible the support was sized to 4 x 8 mesh. The volume of support was measured by loosely packing the material in 250 cc. graduate. The catalysts were reduced with hydrogen at 25 to 100° C. prior to use.

Most of the catalyst supports were washed with distilled water but in the case of the Zeolite 10% sulfuric acid was employed followed by the usual wash with distilled water. The Zeolite-supported catalyst as finally prepared also contained twice as much silver as the other catalysts.

With the exception of the Zeolite these supports were tested only once in this series of experiments. In the initial test with the Zeolite a rather active catalyst was obtained which did not form ethylene oxide but promoted complete oxidation to carbon dioxide. The Zeolite which is used as a water softener, contains a large amount of water-soluble alkali and it was thought that this was probably responsible for the undesirable promoting action on the silver. A second experiment was therefore carried out in which Zeolite was previously treated with 10% sulfuric acid and was coated with a larger amount of silver. This catalyst still had a pronounced tendency to form carbon dioxide but produced appreciable quantities of ethylene oxide.

Plaintiff's Exhibit 28 for Identification (continued).

DISCUSSION:

Although all of the substances tested could be broadly defined as supports a preliminary study would probably eliminate a number of them from further consideration without the need for any experimental work.

In general a support employed in a commercial scale vapor phase process should have certain desirable physical properties including the following:

1. It should be mechanically strong to resist shock and abrasion.
2. It should have a rather rough surface to which the catalyst will adhere well.
3. It should afford a minimum resistance to the flow of gases.
4. It should be cheap and readily available in large quantities.
5. In some cases porosity may be desirable.

It will be noted that among the supports tested are a number having very low mechanical strength and smooth surfaces. A substance such as asbestos would tend to pack tightly and impede the flow of gas. This is still more the case with finely divided materials such as ordinary infusorial earth or precipitated barium sulfate which is why these materials were tested in lump form in these experiments.

Perhaps the most important characteristic of a support is that it should be inert or that any effect should be beneficial. It is known that the materials known as "ceramics" are rather inert chemically. Carborundum, Filtros and Alundum, all of which have been subjected to high temperatures during their preparation, belong to this class of inert supports. Catalytically active forms of alumina and silica such as precipitated aluminum oxide or silica gel are, however, known to have a marked influence on the catalysts in some cases in which they have been employed as supports and might be expected to have some effect on the silver catalyst used in the oxidation of ethylene.

Plaintiff's Exhibit 28 for Identification (continued).

The undesirable action of Zeolite when employed as a support was not unexpected, since alkalis have been widely employed as catalytically active materials.

TABLE III

Support	Source of support	Single Pass Yield %	Efficiency %	Temperature ° C.
Pumice	Eimer and Amend	14.0	51.3	212
Glass	Pyrex, fritted, from Ace Glass Co.	8.8	63.9	200
Brick	Common red building brick	25	51	195
Filtros	Bonded silica, from Filtros Corp.	28	58	206
Carborundum	Bonded carborundum from Carborundum Corp.	16.5	53	212
Porcelain	Unglazed porcelain (Coors) plate from A. H. Thomas Co.	8.4	42	218
Infusorial earth	Infusorial earth bonded with 3% water glass	24.3	43	184
Zinc	Mossy zinc, technical grade from Eimer and Amend	30	37	210
Copper	Copper shot from Eimer and Amend	26.1	42.2	187
Barium sulfate	Barite from Barium Reduction Corp.	21.5	54	211
Magnesium oxide	Calcined magnesite obtained from Eimer and Amend	24.6	53	19[illegible]
Alundum	Bonded aluminum oxide from Norton Co.	27.1	30.6	207
Asbestos	Asbestos fiber, acid and alkali washed, from Eimer and Amend	9	31	207
Zeolite	A synthetic Zeolite, "Decalso", from The Permutit Co.	1.4	9	150
Fire clay	Fire brick	24.8	43	200

Plaintiff's Exhibit 28 for Identification (continued).

EFFECT OF VARYING GAS VELOCITY AND ETHYLENE CONCENTRATION

SUMMARY:

Experiments were carried out to obtain information regarding the effect of varying the concentration of ethylene in the air-ethylene mixture on the production of ethylene oxide. Seven concentrations of ethylene were tested varying from 6 to 80% ethylene.

Experiments were also conducted to determine the effect of varying the gas velocity on the efficiency of the ethylene oxide reaction.

EXPERIMENTAL:

These experiments were carried out in a 1-inch oil jacketed, steel tube heated for a length of 36 inches. Temperatures were recorded through a ¼-inch steel thermocouple well extending through the center of the reaction tube. The catalyst was placed in the lower section of the tube surrounding the thermocouple well and the upper part of the tube was packed with inert material and served as a preheater. The catalyst was prepared by depositing 77 grams of silver oxide on 200 cc. of 4 x 8 mesh carborundum. This silver oxide was then reduced in the tube with hydrogen at 25 to 100°C. The inlet gas was fed at the rate of 50 liters per hour and the temperature was maintained at 250°C. in the concentration experiments. In the velocity experiments the ethylene concentration in the feed gas was about 5% and the catalyst temperature was 260°C.

Additional details and a summary of the results are given in Table I and Table II which are appended.

Plaintiff's Exhibit 28 for Identification (continued).

TABLE I

Effect of Varying Ethylene Concentration

Ethylene Concentration %	Ratio, Air/ Ethylene	Ethylene Oxide Formed mgm/Liter of Exit Gas	Efficiency, %
80	0.25	20.2	46
65	0.52	64.9	63.5
33	2.0	78.4	61
25	3.0	97.7	60
14	6.1	71.9	65
10	9.0	54.1	67
6	15.6	37.8	65

TABLE II

Effect of Varying Gas Velocity

Rate of Feed of Gas, Liters/Hour	Ethylene Oxide Formed mgm/ Liter of Exit Gas	Efficiency, %
200	17.2	63
100	29.7	64
50	52.1	63

j
5-13-40

Plaintiff's Exhibit 28 for Identification (continued).

EFFECT OF ADDING WATER AND CARBON DIOXIDE TO THE INLET GAS

Summary

A series of tests was made in which ethylene was oxidized in the presence of a silver catalyst and of varying amounts of water vapor. The water vapor content of the entering gases was varied from 0.002% to about 3.5%.

The efficiency was not substantially affected by altering the moisture content of the entering gases. In some cases where the drier gases reacted too vigorously the moister gases gave ethylene oxide with better efficiencies. While the water vapor was found to repress the oxidation reactions, in general, the moister entering gases gave as high yields as the less moist gases but usually a somewhat higher temperature was required in the case of the more moist entering gases.

Other tests were carried out in which higher concentrations of water vapor were at times employed. Under certain conditions when dried inlet gases were being used and the temperature rose undesirably the addition of water rapidly restored the normal operating conditions even when the temperature had risen 40 to 50°C. above the optimum and the major portion of the ethylene was being burned to carbon dioxide and water.

Experimental

The catalyst employed was prepared by depositing 77 grams of silver oxide on 200 cc of 4 x 8 mesh Carborundum and reducing the oxide with hydrogen in the reaction tube at 25 to 100°C.

Plaintiff's Exhibit 28 for Identification (continued).

The reaction tube was an oil-jacketed 1-inch tube heated for a length of 36 inches. Temperatures were recorded through a ¼-inch thermocouple well extending through the center of the reaction tube. The catalyst was placed in the lower section of the tube surrounding the thermocouple well and the upper portion of the tube was packed with inert material and served as a preheater. An ethylene-air mixture was fed at the rate of 50 liters per hour to the reaction tube. Mixtures containing 5% and 10% ethylene were employed. Equipment was provided for drying the inlet gas to a dew-point not higher than —55°C. (0.002% water) or for saturating it with water to a dew-point of 26 to 27°C. (approximately 3.5% water). Under the best conditions of the test practically the same single-pass yield, about 45%, and the same efficiency, about 70%, was obtained with both the dried and moist gas but in general somewhat higher operating temperatures were required for the moist gases.

Similar tests made with carbon dioxide added to the inlet gas indicated that the effect of the carbon dioxide was practically the same as that of an equal volume of water vapor.

Tests with another sample of catalyst made in the same way were also carried out in an electrically-heated Pyrex reaction tube having dimensions similar to the steel tube previously described. An ethylene-air mixture containing 5% ethylene and saturated with water vapor at 40°C. (about 7% water) was fed to the reaction tube at the rate of approximately 50 liters per hour. Under normal operating conditions at a temperature of 216°C. ethylene oxide was obtained with a yield of 14.8% and an efficiency of 40%. When dry inlet gas having a dew-point below —55°C. was substituted for the moist feed gas a rapid rise in tem-

Plaintiff's Exhibit 28 for Identification (continued).

perature from 216°C. to 265°C. occurred over a period of ten minutes. Analyses of the exit gas carried out over a temperature range of 240 to 260°C. showed the ethylene oxide was being produced with a yield of 17.9% and an efficiency of only 23.7%. At a reaction temperature of 265°C. and while the temperature was still rising water was added to the inlet gas in such an amount that it formed about 60% by volume of the gas entering the reaction zone. The reaction temperature fell back to the normal operating temperature under these conditions in 10 minutes and operations were resumed with the water vapor concentration decreased to 7%.

6/4/40

Plaintiff's Exhibit 29.

PLAINTIFF'S INTERROGATORIES AND DEFENDANT'S ANSWERS

UNITED STATES DISTRICT COURT
DISTRICT OF MARYLAND

CARBIDE AND CARBON CHEMICALS CORPORATION
Plaintiff,
vs.
U. S. INDUSTRIAL ALCOHOL COMPANY
Defendant.

Civil Action File No. 183

1.

Plaintiff's Interrogatory. Since May 18, 1937, and prior to the filing of the complaint herein, has defendant, or any corporation in which defendant at the time owned and now owns a controlling stock interest, commercially practiced a process of making ethylene oxide by so subjecting a mixture containing ethylene and molecular oxygen at temperatures maintained between about 150° C. and about 400° C. to contact with an active surface catalyst comprising silver for direct oxidation of ethylene by molecular oxygen as to form oxidation products containing substantial amounts of ethylene oxide and not containing substantial amounts of aldehydes, and thereafter recovering ethylene oxide. (By "commercially practiced" is meant—conducted on a commercial scale in the regular course of business and producing a product or products for the purpose of sale and sold in the ordinary channels of trade, as distinguished from experimental or laboratory operations.)

Defendant's Answer. To plaintiff's interrogatory numbered 1, defendant's answer is: No.

2.

Plaintiff's Interrogatory. If the answer to Interrogatory 1 is in the affirmative, by what corporation or corporations and at what place or places was such process so practiced?

Defendant's Answer. To plaintiff's interrogatory numbered 2, no answer is required because defendant's answer to interrogatory numbered 1 is in the negative.

Defendant's Exhibit 46.

B. P. M. Laboratory Report No. 5205 (Portions relied on by plaintiff-appellee in addition to those printed in Defendant-Appellant's Supplement, p. 391 *et seq.*).

* * * * *

On March 30th and 31st a complete test, referred to below as "test 1", was made, an intake gas containing 9.2% ethylene and 18.8% O_2 being practically entirely converted in three cycles without intermediate addition of one of the components. The ethylene content after the third cycle was 0.3%. (When considering this last figure, as also almost all calculations in this report made from gas analyses, it should be taken into account that the *gas analyses* made according to Orsat *are inaccurate to several tenths %*, which, especially with highly diluted gases may result in a *considerable error in the percentage found.* A large number of analyses were made in an endeavor to eliminate this source of error as far as possible).

At my request the demonstration of the first cycle was repeated on April 8th especially with the object of trying to attain complete absorption of the ethylene oxide, whilst also CO determinations were made in the intake and the final gases. This test is referred to below as "test 2".

Appendices 2-5 incl. show in detail the results of the measurements made during said demonstrations, to which may be added the following:

Of course a qualitative reaction test on all products to be formed (ethylene oxide, glycol) was made in all the liquids used in the tests and analyses, but we also tested the absorption and washing liquids obtained for other oxidation products, such as aldehydes, acids, etc. To our great surprise neither the formation of aldehydes, nor of acids or CO was observed, whereas in all publications relating to the direct oxidation of ethylene, formaldehyde,

Defendant's Exhibit 46 (continued).

formic acid and acetaldehyde are mentioned as main or by-products of the partial oxidation (see e. g. *S. Lenher*, J. Am. Chem. Soc. *53*, 3762/1931).

In addition to a *trace of glycol* the only oxidation products found were *ethylene oxide and* CO_2.

* * * * *

For the oxidation of ethylene to ethylene oxide with elementary oxygen the most simple equation of reaction is:

$$2\ C_2H_4 + O_2 \longrightarrow 2\ C_2H_4O.$$

As appears from the numerous publications)1 concerning the oxidation of ethylene, it is very difficult to conduct the reaction in such a manner that a satisfactory yield in *oxide* is obtained. Below certain temperatures ethylene shows little reaction to oxygen, whilst oxidation catalysts readily cause the oxidation to proceed too far. Also high temperatures render it impossible to stop the reaction at a primary oxidation product. However, this primary oxidation product is practically always *formaldehyde*, of which it is easy to understand that at higher temperatures or with active catalysts it undergoes further oxidation or decomposition, the final products of which are *formic acid*, CO, H_2, H_2O and finally CO_2. For conversion into ethylene *oxide* it is necessary to choose such reaction conditions that the double bond of the ethylene is not *broken* up, thus low temperature and very specific catalysts. As, moreover, the rearrangement reaction

ethylene oxide ——— *acetaldehyde*

may give rise to considerable losses of ethylene oxide already formed, it is not surprising that reliable publications describing a process for the satisfactory manufacture of

Defendant's Exhibit 46 (continued).

ethylene oxide (or glycol in the presence of water) by oxidation are very scarce. As far as yields in ethylene oxide are mentioned in these publications, these appear to be inferior to the yield obtained by the S. F. C. G. during the demonstration; we would formulate our conclusion in this regard as follows: *Compared with the other processes so far described for manufacturing ethylene oxide by the reaction of ethylene with oxygen, the S. F. C. G. method gives the best yield, but it will still have to be calculated very carefully whether it is also superior to the methods whereby the oxide is obtained via ethylene chlorhydrine.*

* * * * *

Defendant's Exhibit 55.

SHELL UNION OIL CORP
80 BROAD STREET
NEW YORK

COPY OF MESSAGE
SENT

TO BRANDAN THE HAGUE DATE AUGUST 29, 1933

YOUF 284 REPORTS SHOW C. F. C. G. PROCESS SIMPLE METHOD PRODUCING ETHYLENE OXIDE HOWEVER AT COST DESTRUCTION ONE HALF OF ETHYLENE BY COMPLETE OXIDATION STOP

IF PROCESS APPLIED ON LARGE SCALE MUST PREPARE ETHYLENE AT CONSIDERABLE COST AND DESTRUCTION ONE HALF ETHYLENE BECOMES SERIOUS DISADVANTAGE TO BE BALANCED AGAINST SIMPLICITY IN COMPARING WITH OTHER PROCESSES STOP MOREOVER PATENT POSITION RATHER UNCERTAIN SEE GRATAMS'S LETTERS JULY 29TH AND THIRTYFIRST STOP

THEREFORE MY OPINION OPTION MUST BE EXTENDED FROM AUGUST 31ST TO DECEMBER 31ST TO ENABLE US FURTHER STUDY PROCESS FROM TECHNICAL ECONOMICAL AND PATENT VIEWPOINT BEFORE MAKING RECOMMENDATION FOR AGREEMENT BINDING US TO DEFINITE ROYALTY AND CASH PAYMENT STOP

CONSIDER ANYHOW ROYALTY FAR TOO HIGH AND CASH PAYMENT EXORBITANT BUT CAN ONLY MAKE DEFINITE COUNTER PROPOSALS AFTER COMPLETING STUDY AS MENTIONED ABOVE

D PYZEL

Defendant's Exhibit 63.

Translation

Memorandum on the conferences of Oct. 13 and 14, 1933 concerning the process of the Société de Catalyse Generalisèe, Paris, for the manufacture of ethylene oxide.

Present: Mr. Françon, Soc. de Catalyse Gènèralisèe, Paris

Messrs. Hardonk, Steenackers, Kilian, van Rossum } Bataafsche Petroleum Mij.

Mr. Francon gave us his confirmation that his Société is willing to extend the option for the American patents until Dec. 31, 1933; this will be confirmed by his Société. In the meantime, i. e., until Dec. 31, 1933 the Société de Catalyse Généralisée will not enter into negotiations with other companies concerning the U. S. patents.

* * * * *

Defendant's Exhibit 64.

LETTER dated November 3, 1933, from Masthoff to Jacobson (Portions relied on by plaintiff-appellee in addition to those printed in Defendant-Appellant's Supplement, p. 422).

• • • • •

Geneva's men (whether technical or not I do not know) saw the process exhibited in Paris and were entirely satisfied that the process is workable and that the quoted yields (of 40%) and costs are obtainable.

• • • • •

LVS
11/1/33

Defendant's Exhibit 79.

Postal Telegraph

THE INTERNATIONAL SYSTEM

-N38 74/73 CABLE=PARIS 404P MAR 12 1936

LC DAVIDSON JAYGEDEE=

=NEWYORKNY (DAVIDSON DR J G DAVIDSON CARBIDE AND CARBON CHEMICALS 60 EAST 42 ST RM 1602)=

=FRANCON HAS GIVEN ME NO FIGURE YET FOR SELLING EXCLUSIVE LICENSE HE WILL SEE THE CHAIRMAN PROBABLY SEVENTEENTH MARCH AND GIVE ME HIS TERMS FOR AN OPTION FOR YOU ON ROYALTY BASIS STOP NUMBER AMERICAN PATENT IS 1.998.878 WHICH IS SUFFICIENT TO WORK PROCESS STOP THERE ARE TWO APPLICATIONS OUTSTANDING CONCERNING SIMPLIFICATION IN PREPARATION CATALYST BUT THEY ARE STATED NOT TO BE FUNDAMENTAL AND ARE SECRET FOR PRESENT=

=CHALAS.

1155A

Defendant's Exhibit 91.

JOURNAL OF THE FRANKLIN INSTITUTE

Volume 194, No. 1, July, 1922

"CATALYSIS AND CATALYTIC AGENTS IN CHEMICAL PROCESSES"

by

HUGH S. TAYLOR

(Portions relied on by plaintiff-appellee in addition to those printed in Defendant-Appellant's Supplement, pp. 479 *et seq.*)

* * * * *

The active catalyst is normally a porous, amorphous material and, in process of activation, catalysts tend to assume this form.

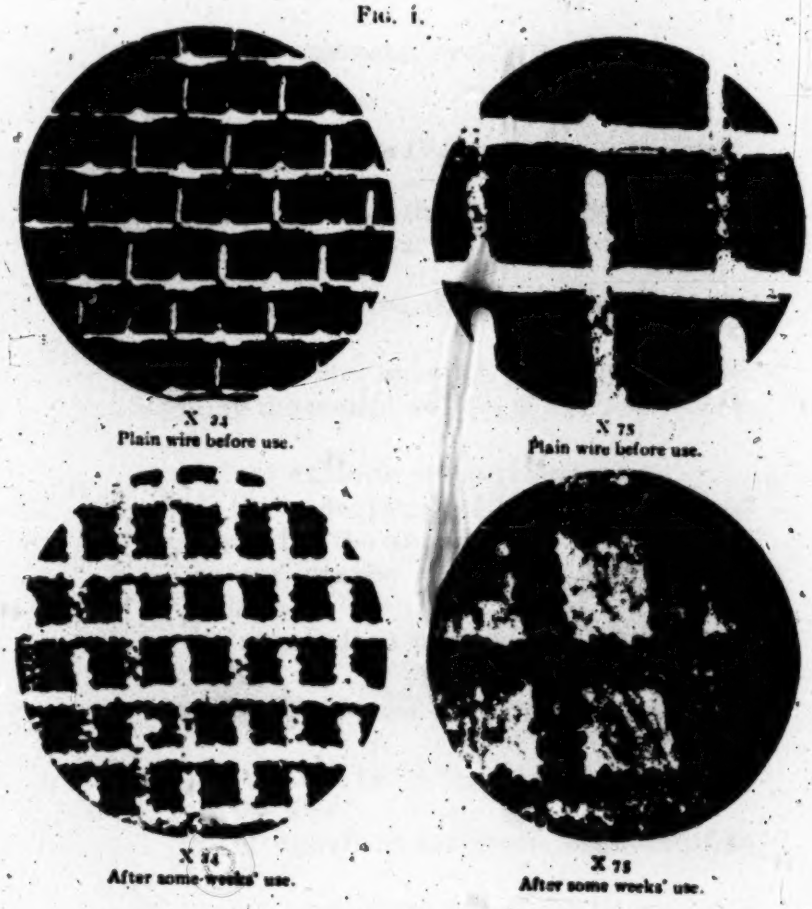
* * * * *

The change is most strikingly manifested in the microscopic examination of platinum gauze before and after use in the oxidation of ammonia. The gauze is converted from a

Defendant's Exhibit 91 (continued).

smooth plane-surfaced wire to a corrugated pitted material. This is shown in the accompanying figure. Similar changes

FIG. 1.



X 24
Plain wire before use.

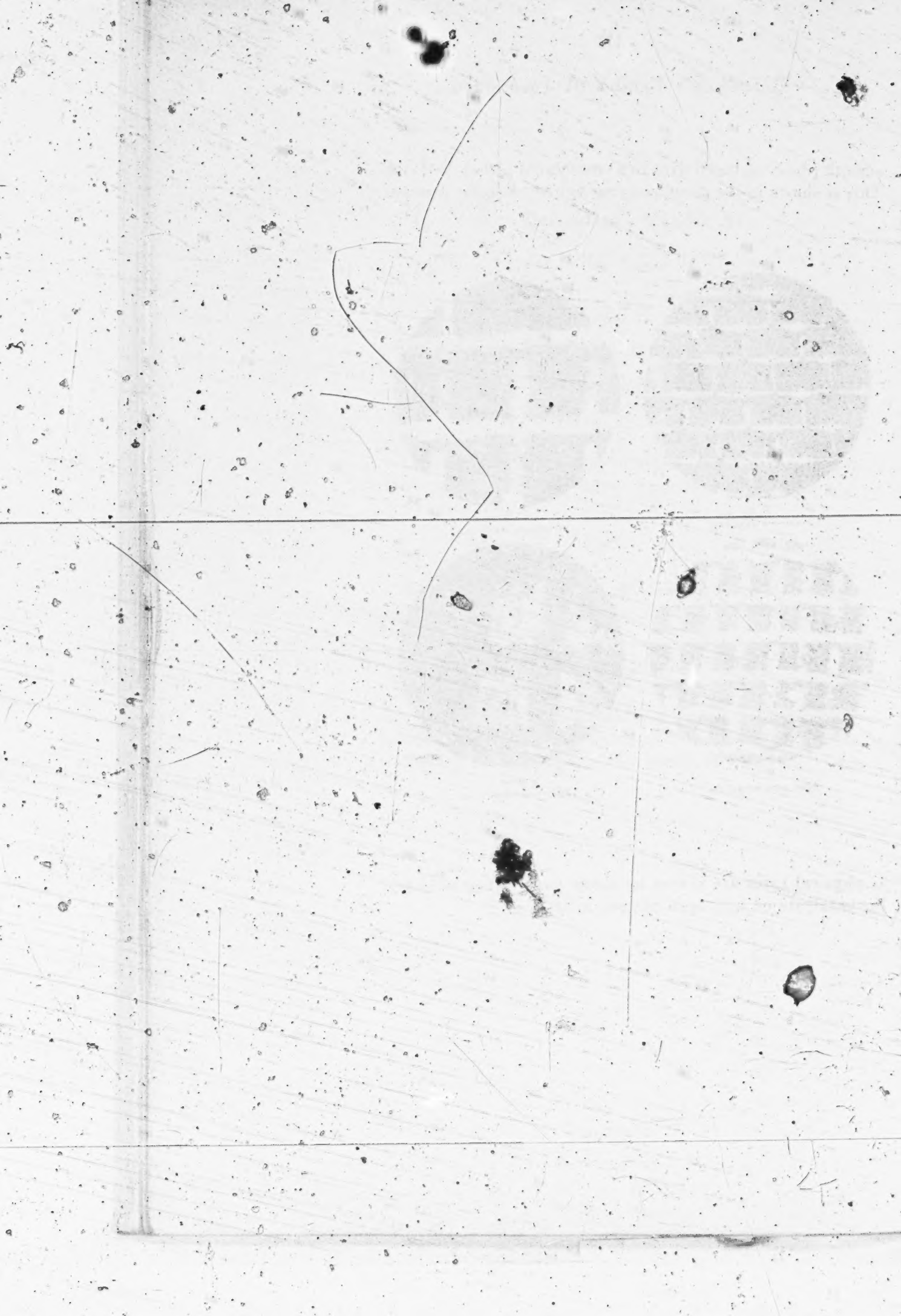
X 75
Plain wire before use.

X 24
After some weeks' use.

X 75
After some weeks' use.

in physical form are shown by silver gauze when used for the catalysts of hydrogen oxygen mixtures.

• • • • •



[fol. 172] PROCEEDINGS IN THE UNITED STATES CIRCUIT COURT OF APPEALS FOR THE FOURTH CIRCUIT

No. 4774

U. S. INDUSTRIAL CHEMICALS, INC., Appellant,

versus

CARBIDE AND CARBON CHEMICALS CORPORATION, Appellee

Appeal from the District Court of the United States for the District of Maryland, at Baltimore

February 26, 1941, the transcript of record is filed and the cause docketed.

Same day, motion and orders extending the time to February 26, 1941, for filing the record on appeal and for docketing the appeal are filed.

Same day, the original exhibits are certified up.

February 28, 1941, the appearance of L. A. Watson and C. V. Johnson is entered for the appellee.

March 1, 1941, the appearance of Dean S. Edmonds is entered for the appellant.

March 3, 1941, the appearance of Harry N. Baetjer is entered for the appellant.

March 17, 1941, plaintiff's exhibit #3 (Operating Model) is certified up.

March 18, 1941, stipulation as to the time for the filing of the respective briefs is filed.

Same day, petition of appellant for leave to file brief in excess of 50 pages is filed.

March 21, 1941, order permitting appellant to file brief in excess of 50 printed pages, but not exceeding 85 printed pages, is filed.

April 1, 1941, brief and supplement on behalf of the ap-
[fol. 173] pellant are filed.

April 9, 1941, petition of appellee for leave to file brief in excess of 50 pages is filed.

Same day, order permitting appellee to file brief in excess of 50 printed pages, but not exceeding 60 printed pages, is filed.

April 11, 1941, brief and appendix on behalf of the appellee are filed.

April 15, 1941, reply brief on behalf of the appellant is filed.

Argument of Cause

April 18, 1941, (April term, 1941) cause came on to be heard before Parker, Soper and Dobie, Circuit Judges, and was argued by counsel and submitted.

[fol. 174] Opinion—Filed June 27, 1941

United States Circuit Court of Appeals, Fourth Circuit

No. 4774

U. S. Industrial Chemicals, Inc., Appellant,

versus

Carbide and Carbon Chemicals Corporation, Appellee

Appeal from the District Court of the United States for the District of Maryland, at Baltimore

(Argued April 18, 1941. Decided June 27, 1941)

Before Parker, Soper and Dobie, Circuit Judges.

Dean S. Edmonds (Venable, Baetjer & Howard; Frank E. Barrows, Charles W. Riley and Frederick L. Bissinger on brief) for Appellant; and Leonard A. Watson (Hershey, Donaldson, Williams & Stanley; C. V. Johnson and C. W. Fairbank on brief) for Appellee.

[fol. 175] Dobie, Circuit Judge:

Carbide and Carbon Chemicals Corporation (hereinafter called plaintiff) brought a civil action in the United States District Court for the District of Maryland against U. S. Industrial Chemicals, Incorporated (hereinafter called defendant), for the alleged infringement of a patent. Judge Coleman, in the District Court, found the material issues in favor of plaintiff and directed that plaintiff recover from defendant profits, gains, advantages and damages, with a reference to a master to ascertain and report to the Court an account of these profits, gains and advantages and to assess the proper damages. Defendant duly took this appeal.

The United States Patent Office granted to Theodore E. Lefort patent #1,998,878 (hereinafter called the original patent), dated April 23, 1935, covering a process for the production of ethylene oxide. Plaintiff was the assignee of the patent. Later, a reissue patent (hereinafter called the reissue patent), Re. #20,370, dated May 18, 1937, was obtained from the United States Patent Office. Plaintiff's claims of infringement relate to the reissue patent and more specifically to Claims #8 and #9 of this patent.

Accordingly, though there are other issues involved in this case, there are two very vital questions that must be considered. The first question involves the validity of the reissue patent, particularly Claims #8 and #9. The second question, if such validity be sustained, is concerned with the infringement by the defendant.

The Validity of the Reissue Patent

The point most seriously advanced by defendant, both in the briefs and in oral argument, is the invalidity of the reissue patent, particularly Claims #8 and #9, on the ground that the process disclosed in the reissue patent involves a substantial and material (even an essential) va- [fol. 176] riance from the process set out in the original patent. More precisely, defendant claims that, in the process for the production of ethylene oxide under the original patent, the voluntary addition of water into the reaction-chamber is not only mandatory but is an essential step in this process; while under the reissue patent, the process calls for no voluntary introduction of water into the reaction-chamber. It is absolutely clear that, with or without this voluntary introduction of water, some water is, and must be, there formed through a side-reaction in the process.

The law is clear, under the reissue statute (Rev. St. § 4916; U. S. C. A., Title 35, § 64), that there can be no reissue patent for a new invention. Indeed, the cases go much further than this to hold that the reissue patent is invalid if, between the inventions disclosed first in the original patent and then in the reissue patent, there be variances that are essential, substantial and material. A leading case is *Parker & Whipple Co.* v. *Yale Clock Co.*, 123 U. S. 87, —— S. Ct. ——. Other important cases are *Seymour* v. *Osborne*, 11 Wall. 516, —— S. Ct. ——; *Powder Co.* v. *Powder Works*, 98 U. S. 126, —— S. Ct. ——; *Mahn* v.

Harwood, 112 U. S. 354, —— S. Ct. ——. And the United States Supreme Court has cited and applied the *Parker & Whipple Co. case, supra*, in *Matthews* v. *Ironclad Mfg. Co.*, 124 U. S. 347, 351, —— S. Ct. ——; *Yale Lock Co.* v. *James*, 125 U. S. 447, 464, —— S. Ct. ——; *Corbin Cabinet Lock Co.* v. *Eagle Lock Co.*, 150 U. S. 38, 42-43, —— S. Ct. ——. And see, decided by this Court, the cases of *Marvel Buckle Co.* v. *Alva Mfg. Co.*, 196 F. 1006; *A. D. Howe Machine Co.* v. *Cooffield Co.*, 197 F. 541. Yet a reissue patent is not invalid merely because the reissue patent contains claims broader than the original, *Topliff* v. *Topliff*, 145 U. S. 156, —— S. Ct. ——, or claims narrower than the original, *Miller* v. *Brass Co.*, 104 U. S. 350, —— S. Ct. ——.

In the instant case, the original patent (all italics are [fol. 177] ours) thus begins: "This invention has for object a process for the production of ethylene oxide which mainly consists in subjecting ethylene to the simultaneous action of the oxygen of air and of *water* in presence of a catalyzer and, if need be, of hydrogen." Later, the patent (page 1, column 2, lines 10-11) states: "*Water* can be *admitted* (not formed or developed) in the reaction vessel, either in the liquid state, or as steam." Also we find (page 1, column 2, lines 15-16) "*introduction*, (again a word of conscious action) in this reaction, of a suitable quantity of *water*." Again (page 1, column 2, lines 36-43) is: "if *water* is *introduced* in suitable quantity, the reaction is not only facilitated, as above stated, but, in addition, the reaction giving CO_2 (carbon dioxide) * * * is checked." (Words in parentheses are interpolated).

Three modi operandi are set out in the original patent. In the first, there is (page 2, column 1, line 18) "a circulating pump allowing to *introduce water*", and (page 2, column 1, lines 20-21) "The reaction will take place in this tube between ethylene, the oxygen of air, and water." In the second (page 2, column 1, lines 33-35), is "a mixture of ethylene, air, *water* vapour, and hydrogen is *caused to pass* therethrough (through the tube)." And, in the third (page 2, column 1, lines 41-44), is: "A silver colloid is introduced in a high pressure tube which *has been filled* with *water*. Pure ethylene under pressure is *then* added in order that it can dissolve in the water"; and (page 2, column 2, lines 3-5): "*Water*, in the form of steam or not, *considerably promotes the reaction ensuring* the production of ethylene oxide."

Not so strong (as to the voluntary introduction of water into the reaction chamber) is the language in the original patent of the seven claims. The first three claims *each* begin (page 2, column 2): "A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and *water*." In claims 3 and [fol. 178] 4 (page 2, column 2) the language is identical, save that the word "steam" (a form of water) is substituted for the word "water". While in claims 6 and 7 (page 2, column 2) after the word "oxygen", we find the words "steam and hydrogen".

Apart from this verbal analysis of the original patent, we have made a careful study of the record. We are convinced that this original patent contemplated the voluntary introduction of water into the reaction-chamber, not as an optional procedure, but as a step that would always be taken. The verbal analysis shows that, in the original patent, literally there is "Water, water, everywhere". Nor is it water developed as a by product, but it is water voluntarily introduced into the tube to bring about the desired reaction. There is some expert testimony in the record which might seem to prove the contrary. This testimony, *as to the specific point now before us*, is considerably weakened by two considerations. These experts were super-chemists of unusual distinction; they were not the patentee Lefort. They drew not only upon their vast store of technical knowledge, but they used, too, what developments (and these were far from inconsiderable) had been made in the whole field of the Lefort patent since application for this patent was made by the inventor. Some study of chemical history and literature, as these existed at the time of Lefort's labors, convinces us even more strongly of the truth of the conclusion that the voluntary introduction of water was a mandatory provision of the original Lefort patent.

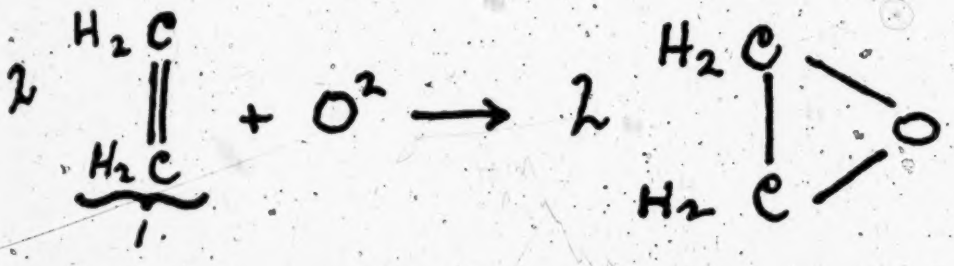
It seems quite clear, on the other hand, that the reissue patent does *not* contemplate the voluntary introduction of water into the reaction-chamber as a necessary step in the procedure. There is no mention of the word "water" in the introductory paragraphs of the reissue patent (apart from an incidental statement on page 1, column 1, line 23, that oxidation of ethylene gives some carbon dioxide and [fol. 179] water). Indeed, the word "water" is not found until the 39th line of page 1, column 1, and note how different

is the language here used, when compared with the language of the original patent. Here we find (page 1, column 1, lines 39-42): "*Some water* or carbon dioxide in addition to that formed *can* be admitted to the mixture in the reaction zone if desired." And the word "water" occurs only once more in the reissue patent before the claims are set out. We find (page 1, column 2, lines 12-16): "Ethylene and oxygen are thus reacted simultaneously at the temperatures set forth *in the presence of a surface catalyst and water.*" Claims #1 to #7 in the reissue patent present no important differences from the like-numbered claims in the original patent. Added Claims #8 and #9 (the claims at issue in the instant civil action) each use the word "water" but once, in the expression "to form oxidation products containing ethylene oxide *in the presence of water*" (Claim #8, page 2, column 1, line 35, and Claim #9, page 2, column 2, line 25). In reference to the use of "water", such verbs as "admitted" or "introduced", with the single exception first discussed above when the term "admitted" is most carefully qualified and restricted, are conspicuous by their absence from the reissue patent.

This brings us to what we regard as the most crucial problem in the case. Are Claims #8 and #9 of the reissue patent so vitally and so substantially different from the original patent as to invalidate these claims? We have just held that *there is a difference* here in that the original patent contemplated the voluntary introduction of water into the reaction-chamber (distinguished from the mere formation of water during the course of the reactions) as a step that would always be taken in the procedure therein set out, while Claims #8 and #9 contain no such requirement, and indeed seem to negative clearly any such step. Is this difference vital, essential, substantial, material? [fol. 180] Judge Coleman answered this question categorically in the negative, in his findings of fact number 7 and number 8. Under Civil Rules of Federal Procedure, Rule 52(a), we can set aside these findings if, but only if, in our opinion the findings are "clearly erroneous". We do not think these findings are "clearly erroneous", so we must affirm them.

The Lefort process was one for the production of ethylene oxide C_2H_4O, by a direct chemical combination between ethylene, C_2H_4, which is a hydro-carbon gas, and oxygen. The chemical formula for this conversion, as set

out in the original patent (page 1, column 1, lines 11-14), is thus written:



This means that 2 molecules of ethylene plus molecular oxygen (2 atoms) will produce 2 molecules of ethylene oxide. This chemical formula had long been known. The difficulty was to find some practical process for bringing about the reaction prescribed by the formula. Some eminent chemists had stated that no such process would ever be found. The process described in Claims #8 and #9 of the reissue patent does constitute such a process, and there is no doubt of the real practical importance and the great commercial value of this process.

We think it is fair to state, in the light of the record in this case, that under the original patent, the essentially vital aspects of the process are (1) subjecting ethylene to the action of the oxygen of air; (2) in a reaction-chamber at controlled and specified temperature; (3) in the presence of a designated type of catalyst. The role of the water deliberately introduced into the reaction chamber was to increase and facilitate the efficiency of the reaction [fol. 181] by diminishing the quantities of carbon dioxide and other undesirable by-products which might result from the reaction. Water would furnish a cheap and available heat-absorbing diluent. Lefort, of course, knew of the presence of some moisture in air. However, the original patent does not mention the quantity or proportion of water that was to be voluntarily introduced. There is mention, in the third modus operandi, of a "tube which has been filled with water"; but just before the setting out of the three modi operandi, we find the statement: "Several modi operandi for carrying the process into practice, will be indicated hereinafter, by way of example only." The specifications of the original patent indicate the introduction "of a *suitable quantity* of water" (page 1, column 2, lines 15-16) and "if water is introduced in suitable quantity" (page 1, column 2, lines 35-36).

There are a very few differences between Claims #8 and Claim #9 of the reissue patent, and these differences are hardly of very great importance. Claim #8 mentions "a mixture containing ethylene and molecular oxygen"; Claim #9 adds to this the words "and a heat-absorbing diluent". Claim #8 mentions a "catalyst of a material having the catalytic characteristics of silver". In Claim #9 we find "catalyst material composed essentially of silver". Claim #8 ends "and thereafter recovering ethylene-oxide-containing product in liquid form". Claim #9 concludes "and thereafter separating ethylene oxide from the products of oxidation."

Claim #8 of the reissue patent is reproduced in full, with the numerals therein inserted by Judge Coleman to indicate the language covering the six steps embraced in the process:

"In both claims 8 and 9, the process set forth for the production of ethylene oxide embraces six steps. We quote claim 8 in full, and have inserted numerals indicating the language in the claim which covers these steps: 'The process of making ethylene oxide by the direct chem- [fol. 182] ical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene, which comprises (1) forming a mixture containing ethylene and molecular oxygen and (2) conducting said mixture through a confined reaction zone which is maintained at an elevated temperature; (3) controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400° C.; (4) subjecting said mixture in said zone at said elevated controlled temperature to intimate contact with an active surface catalyst of a material having the catalytic characteristics of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water, and which does not favor the existence in said products of substantial amounts of aldehydes; (5) correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide; and (6) thereafter recovering ethylene oxide-containing product in liquid form.'"

This process is thus described by Judge Coleman in his opinion:

"Shorn of technical phraseology and summarized very briefly, the process may be described as consisting of the passage of a mixture of ethylene gas and air through a tube containing a silver catalyst heated to a control temperature between approximately 150° and 400°. The direct combination of the air with the ethylene produces ethylene oxide. A side reaction also occurs in that there is destructive oxidation of some of the ethylene, producing carbon dioxide and water. It is claimed in the introductory statements of the patent that 'This process is characterized by an entirely unique reaction in which the direct addition of a single oxygen atom to the ethylene molecule occurs without the formation of significant amounts of other ox-genated compounds such as aldehydes.' That is to say, the product is claimed to be a notably clean one, —pure ethylene oxide, in readily recoverable form."

The expert testimony here strongly favors the conten- [fol. 183] tions of the plaintiff. Professor Lamb, after a series of experiments, testified that one skilled in the art would, by following the original patent, successfully produce ethylene oxide, whether the introduction of water be regarded as mandatory or optional, and that he would obtain the same practical result by following the teachings of the reissue patent. In no uncertain terms, he gave his opinion, based upon his experiments conducted before he had ever seen the reissue patent, that, if it be assumed that the original patent required the introduction of water and the reissue patent omitted this requirement, this, from the technical viewpoint of chemistry, would not constitute a material or substantial variance. Professor Lamb remained at the trial and was recalled and asked by Judge Coleman: "Have you, since you testified, had a chance to study the reissue patent?" Professor Lamb replied: "Last evening I read it for the first time, so I have some familiarity with it." To the court's question "Well, then, do you feel you are in a position to testify more fully now from a scientific standpoint as to whether you find any material difference on this question that has been dwelt upon so much, the departure, in that water is required in the first patent and not in the second, as an addition?", Professor Lamb's reply was: "I certainly have not heard anything

that has changed my ideas about water as an ingredient in this reaction."

The testimony of Professor Lamb was strongly confirmed by the evidence of Professors Frazer and Stevenson and of Dr. Curme, director of research for plaintiff. Professor Frazer stated emphatically: "In my judgment, the amount of water or the manner in which it was added was not the important thing at all. Speaking entirely as a chemist." Again, he stated: "As far as I could make out in regard to this question of water, it is a very much over-worked topic."

The testimony of the experts introduced by defendant, [fol. 184] we think, does not bear favorable comparison with the testimony of the experts introduced by plaintiff. Nor was defendant helped by virtue of the fact that Judge Coleman appointed Professor Frazer as an inter-partes expert witness in order that experimental tests might be made in advance of the trial and defendant produced no witness who had made, or who co-operated with Professor Frazer in making any inter-partes tests along the lines of either the original patent or the reissue patent.

Two points have been suggested to us, which, it is said, should heavily discount the value, in this connection, to the plaintiff of its expert witnesses in general, and of Professors Lamb and Frazer particularly. These are not merely ordinary men "skilled in the art". See U. S. C. A. Title 35, Section 33. They are superior supermen in the science of chemistry, to whom, in the light of their vast knowledge, the specifications and claims of a patent suggest possibilities that did not occur to the patentee and would not occur to the ordinary skilled man. Again, these super chemists are primarily interested in the scientific chemical possibilities of a patent; but they have little interest or knowledge in the patent law and the monopoly thereby granted to an inventor, or in practical commercial considerations such as the fact that if water, here, be voluntarily introduced into the reaction-chamber in appreciable quantity, the expense and trouble involved in heating this water to the desired temperature would materially enhance the costs of production. We have given due weight to these suggestions which, in our opinion, have some unquestioned force. Yet, even after such weight has been given, we still think their testimony, set against, and read

in the background of, the entire record of the instant case, affords ample warrant for the findings by Judge Coleman.

Defendant contends that the language of the patents here in suit is addressed to, and must be interpreted by, "persons skilled in the art or science to which it appertains" (U. [fol. 185] S. C. A. Title 35, Section 33), rather than by such super-chemists as the expert witnesses who testified for plaintiff. In *Seymour* v. *Osborne*, 11 Wall. 516, at pages 545-546, we find:

"Whether a reissue patent is for the same invention as that embodied in the original patent or for a different one is a question for the court in an equity suit to be determined as a matter of construction, on a comparison of the two instruments, *aided or not by the testimony of expert witnesses, as it may or may not appear that one or both may contain technical terms or terms of art requiring such assistance in ascertaining the true meaning of the language employed.* * * *

Cases doubtless arise where the language of the specification and claim, both of the surrendered and reissued patents, is so interspersed with technical terms and terms of art that *the testimony of scientific witnesses is indispensable to a correct understanding of its meaning.*" (The italics are ours).

The very field of the instant patents, catalytic chemistry, is a highly specialized one in which there have been developments that are many and important. The question of substantial variance *vel non* between the two patents, original and reissue, is close and troublesome due in no small measure to the difficulties involved not only in analytically interpreting and applying individual words of a highly technical nomenclature, but also in mentally envisaging complex chemical processes in their entirety and in accurately and fairly comparing these processes when so envisaged. We, accordingly, think our case falls squarely into the category described in *Seymour* v. *Osborne*.

Defendant attempted to make much of other French and British patents, which, like a scarlet thread, run through defendant's brief. Any adequate discussion of these foreign patents, in this connection, would unduly prolong this opinion. In answer to defendant's contention, however, that the reissue was largely an embodiment of the

[fol. 186] details set out in these foreign patents, it may be pointed out that the lawyer who had charge of the application for the re-issue patent testified, without contradiction or impeachment, that he then had no knowledge whatever of the details of these foreign patents.

The specifications of the original patent, at the most, did not add to the elements called for by the claims, but merely contained an inadvertent error as to one step in the process (i. e., the voluntary introduction of water), and that not one of the most important steps, in the process by which these elements were to be brought into functional relation. A reissue patent would seem to be a proper method for the correction of such an error. And, if claims #8 and #9 of the reissue patent had been added to the claims of the original patent, we do not believe these claims could have been held to be invalid on the ground that these claims were not sufficiently covered by the specifications of the original patent.

Minor Defenses

We regard the issue of a substantial variance between the original patent and the reissue patent as the vital question involved in this appeal. Having resolved that question in favor of the plaintiff, we now proceed to certain minor defenses set up by defendant. Judge Coleman, in his opinion, considered and dealt with these minor defenses at some length. He decided that all of them were lacking in merit. We believe all these rulings to be correct and the reasons on which they were based to be sound. We shall advert somewhat briefly to each of these minor defenses.

Defendant attacks the validity of the original patent on the ground that under the prior art this patent lacks patentable novelty. Even a cursory reading of the testimony of Professor Lamb, Professor Stevenson and Dr. Curme should effectively silence this contention. The Lefort [fol. 187] process provided a practical and original solution to a problem that had long baffled industrial chemists. We certainly do not believe that the original patent was anticipated by the McElroy patents, for the McElroy patents were demonstrated to be quite unworkable in any practical commercial sense, and these patents fail to disclose the essential features of Lefort's original patent.

The federal Reissue Statute (59 Rev. St. §4916; 35 U. S. C. A. §64) permits a reissue patent only if error in an original patent "has arisen by inadvertence, accident or mistake, and without any fraudulent or deceptive intention." The evidence amply sustains the finding of the United States Patent Office (See *Topliff* v. *Topliff*, 145 U. S. 156, —— S. Ct. ——), affirmed by Judge Coleman, that the conditions set out in the Reissue Statute have been adequately satisfied here. There is little force in defendant's contention that the claims and specifications of the original patent were fraudulently or deceptively made vague and indefinite. Unusually convincing in this connection, are the testimony of plaintiff's attorney and the affidavit of the French attorney. This seems to be a typical case of a valid, original basic patent, in which the claims were inadvertently drawn in terms that were too narrow and these claims were, accordingly, subject to valid correction under a reissue patent.

Another contention is that Claims #8 and #9 of the reissue patent specify the catalyst only in terms of function. The testimony of Professor Lamb disposes of this contention. He was amply confirmed here by Professor Stevenson's evidence and particularly the testimony of the latter dealing with the literature of silver catalysts, including the notable contributions on that subject of Professor Benton (a catalytic chemist of outstanding distinction) and Mr. Elgin. Nor was there much encouragement to defendant here in the testimony of its own expert, Mr. Ray Carter. We have no reasonable doubt that the description of the catalyst would be entirely sufficient to [fol. 188] enable one versed in the art to follow satisfactorily the outlined process.

Defendant asserts that plaintiff has failed to disclaim claims #2, #4, #5, #7 of the reissue patent, though the invalidity of these claims was brought to the attention of the plaintiff in the answer of the defendant, and that this fact makes the reissue patent invalid in its entirety. These claims have never been adjudged to be invalid, nor do we think their invalidity is clearly apparent upon an inspection of the patent. The approval of these claims by the Patent Office would seem to raise some presumption, until the contrary appears, that the claims are valid. It is at least open to very serious doubt whether "the making of the invalidity of these claims obvious" by a mere asser-

tion to that effect in its answer would relieve the defendant from liability for infringement of claims #8 and #9 committed prior to the filing of this answer. Claims #8 and #9, (not the narrower claims #2, #4, #5, #7) form the basis of the instant action.

Said Judge Coleman on this point: "A patent will not be invalidated for mere failure on the part of the patentee to disclaim prior to some judicial decision upon the subject matter, i. e., upon one or more of the claims of the patent. After that, the patentee must act with diligence in accordance with the result of the adjudication." And in Robinson on Patents (1890) vol. 2, page 284, we find the following language, cited with approval in *Easter* v. *Simon, Ascher,* 282 U. S. 445, 453: "In cases where the excess is not apparent at once upon the inspection of the patent by the patentee, the allowance of his claim by the Patent Office raises such a presumption in his favor that he may rely upon its validity until a court of competent jurisdiction decides that it is broader than his real invention." The cases of *Easter* v. *Simon, Ascher,* 282 U. S. 445; *Triplett* v. *Lowell,* 297 U. S. 638; *Maytag Co.* v. *Hur-*
[fol. 189] *ley Machine Co.,* 307 U. S. 242, *Radio Condenser Co.* v. *General Instrument Corp.,* 65 F. (2d) 458 (cited in the brief of defendant); do not go beyond what is written above. Hence, they do not sustain the interpretation sought to be placed upon them by the defendant.

If Claims #8 and #9 of the reissue be valid (and we do so hold), we think there can be little doubt of infringement by defendant. Indeed, we do not believe defendant's contention to the contrary was very seriously urged. The deadly parallel between the process of the reissue patent and the process employed by defendant, set out in plaintiff's exhibits #8 and #9, which was not successfully contradicted by defendant, is quite sufficient here.

Defendant's last defense is in the nature of an attempt to invoke against plaintiff an equitable estoppel, based largely on expenditures by defendant before the issuance of the reissue patent. Defendant is here clutching for a very feeble straw. *Sontag* v. *National Nut Co.,* 310 U. S. 281, —— S. Ct. ——, and *Otis Elevator Co.* v. *Atlantic Elevator Co.,* 47 F. (2d) 545, are clearly distinguishable on the facts from the instant case. The expenditures in question were largely for experimental purposes. Even as to these, the evidence is far from clear as

to just how much money was expended before, and just how much after, the application for the reissue patent. As Judge Coleman observed:

"But it appears that defendant never began to construct its present costly plant until after the reissued patent had been granted; and never began to operate it commercially for the production of ethylene oxide until more than two years later. It is a correct inference from the testimony that prior to the application for the reissued patent, defendant's work on this process amounted to relatively little, but that thereafter, defendant zealously progressed its development of plant and process, with a view to capitalizing what plaintiff had done."

[fol. 190] *General Considerations*

We believe that what might be called the general equities here strongly favor the plaintiff. A French inventor, Lefort, made a discovery of great scientific importance and unquestioned commercial value. Plaintiff acquired the original patent, which was perfected by a reissue patent. It cannot fairly be said that plaintiff merely bought a lawsuit or tried to play the role of a dog in the manger. And this is true, even though there may have been some doubt as to the validity of the patents in question.

On the other hand, the activities and expenditures of defendant in connection with ethylene oxide were comparatively slender before application was made for the reissue patent. The erection of defendant's elaborate plant in Baltimore after the issuance of the reissue patent, and defendant's elaborate activities thereafter were all in the nature of a gamble on the invalidity of the reissue patent by the defendant's officers, who hoped, if they were thus fortunate, to prosper by the appropriation of the intellectual property of another. If, through over-hasty action, this chance broke against defendant, we see little need for the shedding of any crocodile tears over the misfortune thus brought on the defendant.

Finally, we are impressed by the general nature of the defenses set out at length by the defendant in a desperate effort to escape the consequences of a palpable infringement. These defenses, terribly technical in the extreme, contain little or no appeal to the beneficent principles of equity and good conscience.

For the reasons set forth above, we affirm the judgment of the District Court.

Affirmed.

[fol. 191] DECREE—Filed and Entered June 27, 1941

UNITED STATES CIRCUIT COURT OF APPEALS, FOURTH CIRCUIT

No. 4774

U. S. INDUSTRIAL CHEMICALS, INC., Appellant,

vs.

CARBIDE AND CARBON CHEMICALS CORPORATION, Appellee

Appeal from the District Court of the United States for the District of Maryland

This Cause came on to be heard on the transcript of the record from the District Court of the United States for the District of Maryland, and was argued by counsel.

On Consideration Whereof, It is now here ordered, adjudged, and decreed by this Court that the decree of the said District Court, in this cause, be, and the same is hereby, affirmed with costs.

June 27, 1941.

Armistead M. Dobie, U. S. Circuit Judge.

July 22, 1941, petition of appellant for stay of mandate and for extension of time to present petition for rehearing is filed.

July 26, 1941, order extending time to August 27, 1941, within which to present petition for rehearing, and staying mandate, is filed.

August 19, 1941, petition of appellant for a rehearing is presented.

[fol. 192] ORDER DENYING REHEARING—Filed and Entered September 2, 1941

UNITED STATES CIRCUIT COURT OF APPEALS, FOURTH CIRCUIT

No. 4774

U. S. INDUSTRIAL CHEMICALS, INC., Appellant,

versus

CARBIDE AND CARBON CHEMICALS CORPORATION, Appellee

In the above entitled cause it is ordered that the petition for rehearing presented August 19, 1941, be and the same is hereby denied.

Sept. 1st, 1941.

John J. Parker, Senior Circuit Judge; Morris A. Soper, United States Circuit Judge; Armistead M. Dobie, United States Circuit Judge.

September 5, 1941, petition of appellant for a stay of the mandate is filed.

ORDER STAYING MANDATE—Filed September 8, 1941

[Style of Court and Title Omitted]

Upon the Application of the appellant, by its counsel, Dean S. Edmonds, and for good cause shown,

It Is Ordered that the mandate of this Court in the above entitled cause be, and the same is hereby, stayed pending the application of the said appellant in the Supreme Court of the United States for a writ of certiorari to this Court, unless otherwise ordered by this or the said Supreme Court, and provided said application is filed in the said Supreme Court within 30 days from this date.

September 6, 1941.

John J. Parker, Senior Circuit Judge.

[fol. 193] STIPULATION AS TO RECORD FOR USE IN SUPREME COURT ON APPLICATION FOR WRIT OF CERTIORARI—Filed September 16, 1941

UNITED STATES CIRCUIT COURT OF APPEALS, FOURTH CIRCUIT

No. 4774

U. S. INDUSTRIAL CHEMICALS, INC., Appellant,

vs.

CARBIDE AND CARBON CHEMICALS CORPORATION, Appellee

It is hereby stipulated by and between counsel for the respective parties hereto that the record to be certified by the Clerk of the Circuit Court of Appeals for use in the Supreme Court on the application for certiorari shall consist of the parts of the record contained in the appendix to the brief of the appellant and the appendix to the brief of the appellee, together with the proceedings in the Circuit Court of Appeals.

It is further stipulated that, in addition to the above transcript, the Clerk shall make up and send to the Supreme Court a complete transcript of the record for use in that court, if desired or required.

Dean S. Edmonds, Of Counsel for Appellant. L. A. Watson, Of Counsel for Appellee.

Dated: September 11, 1941.

[fol. 194] CLERK'S CERTIFICATE

UNITED STATES OF AMERICA,
Fourth Circuit, ss:

I, Claude M. Dean, Clerk of the United States Circuit Court of Appeals for the Fourth Circuit, do certify that the foregoing three (3) volumes is a true copy of the Supplement to Appellant's Brief; Appendix to Appellee's Brief, and of the proceedings in the said Circuit Court of Appeals in the therein entitled cause, as the same remain upon the records and files of the said Circuit Court of Appeals, and constitute and is a true transcript of the

record and proceedings in the said Circuit Court of Appeals in said cause, made up in accordance with the stipulation of counsel for use in the Supreme Court of the United States on an application for a writ of certiorari.

In Testimony Whereof, I hereto set my hand and affix the seal of the said United States Circuit Court of Appeals for the Fourth Circuit, at Richmond, Virginia, this 24th day of September, A. D., 1941.

Claude M. Dean, Clerk, U. S. Circuit Court of Appeals, Fourth Circuit. Seal.

[fol. 195] SUPREME COURT OF THE UNITED STATES

U. S. INDUSTRIAL CHEMICALS, INC., Petitioner,

vs.

CARBIDE AND CARBON CHEMICALS CORPORATION, Respondent

STIPULATION

It Is Hereby Stipulated By Counsel that the Petition for Certiorari may be considered on the appendices to the briefs of the parties as supplemented by the Circuit Court of Appeals proceedings.

It Is Further Stipulated that the original record be lodged with the Clerk and that in the event the Petition for Certiorari is granted counsel will then stipulate to print from the original record those parts which they deem pertinent.

Dean S. Edmonds, Counsel for Petitioner. L. A. Watson, Counsel for Respondent.

Dated: New York, N. Y., September 26, 1941.

[fol. 196] SUPREME COURT OF THE UNITED STATES

ORDER ALLOWING CERTIORARI—Filed November 10, 1941

The petition herein for a writ of certiorari to the United States Circuit Court of Appeals for the Fourth Circuit is granted.

And it is further ordered that the duly certified copy of the transcript of the proceedings below which accompanied the petition shall be treated as though filed in response to such writ.

[fol. 197] SUPREME COURT OF THE UNITED STATES

STIPULATION AS TO RECORD—Filed December 3, 1941

The petition for certiorari herein having been considered by the Court on the appendices to the briefs of the parties as filed in the Court of Appeals for the Fourth Circuit supplemented by the proceedings in the Court of Appeals, and the original record herein having been lodged with the Clerk of this Court, and the said petition for certiorari having been granted and the writ of certiorari issued,

It Is Hereby Stipulated by counsel that this case may be considered by the Court on the return of the writ of certiorari on the record as presented by the said appendices as filed by the parties in the Court of Appeals for the Fourth Circuit, and the physical exhibits there introduced.

Dan S. Edwards, Counsel for Petitioner. Leonard A. Watson, Counsel for Respondent.

Dated: New York, N. Y., December 1, 1941.

[fol. 198] [File endorsement omitted.]